Screening of biogas methanation in Denmark

Resources, technologies and renewable energy integration

Skov, Iva Ridjan; Nielsen, Steffen; Nørholm, Malte Skovgaard; Vestergaard, Johann Pálmason

Publication date:
2019

AALBORG UNIVERSITET

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April, 2019

Iva Ridjan Skov

Steffen Nielsen

Malte Skovgaard Nørholm

Johann Pálmason Vestergaard

Aalborg University
Department of Planning

Publisher:
Department of Planning
Aalborg University
Rendsburggade 14
9000 Aalborg
Denmark

ISBN 978-87-93541-09-2

Abstract

This report provides an overview of existing biogas resources and biogas production in Denmark. The analysis includes mapping of manure, straw and municipal waste across municipalities in the country. Furthermore, it presents research and development of biogas upgrading and biogas methanation technologies at existing plants including the status of electrolysis technologies. The potential for renewable energy integration was analysed for 3 Danish scenarios: reference 2020 as well as 2035 and 2050.

We regard biogas methanation as one of the key technologies in future renewable energy systems.

This report is prepared as a part of Task 2.5 in the EUDP Biocat Roslev project

CONTENT

SUMMARY

The biogas production in Denmark has increased by more than 55% from 1980 to 2017, where the biogas production reached 11.16 PJ. From 2015 to 2017, the biogas production has increased by 44% and it has increased further in 2018. In the last 6 years, the number of biogas upgrading plants have increased from 6 to 33 plants that deliver methane to the gas network. Denmark has become a mature market for biogas upgrading technologies. Biogas methanation in Denmark has gained interest in the last couple of years, with currently 3 demonstration plants in operation; two with biological methanation and one with catalytic methanation.

This report shows the biogas resource potential by mapping manure and bedding, straw and biodegradable municipal waste in all Danish municipalities. The results show that the biogas potential for manure and bedding is 27,632,435 tons and 2,315,437 tons (incl. dry matter), respectively. For straw (8 most common types), the potential is 3,728,967 tons (incl. dry matter) and for biodegradable waste, it is 2,960,387 tons.

Denmark is rich in biomass resources per capita, making the biogas potential high. The methane potential from biogas in Denmark ranges from 32 PJ to 107 PJ, including electro-methane from biogas methanation with electrolytic hydrogen based on different sources. This means that, in the future, the role of biogas methanation could be high depending on the resources actually available. In this report, the total potentials for electro-methane are 25.56 PJ in 2020, 33.5 PJ in 2035 and 55 PJ in 2050, assuming that the full biogas potential is methanised with the addition of hydrogen.

While biogas upgrade is a rather mature technology and has a variety of processes that can be used for this purpose, the biogas methanation technology is emerging. The production of electrolytic hydrogen from alkaline electrolysis is the most mature process; however, it shows limitations to dynamic operation if operated under atmospheric pressure and on large scale. PEM electrolysis is now a commercially available technology that is getting more widespread on the market due to its flexible operation. SOEC is still in the development phase and the technology is yet to be commercialized. Once hydrogen is produced, the methanation of the carbon dioxide part of biogas takes place. Catalytic methanation is a commercialized process, while biological methanation only recently has reached a commercial level. The upscaling of the technology is the next step towards the large-scale implementation of power-to-gas (P2G) via biogas methanation. This report includes the state-of-the-art of biogas upgrading, biogas methanation and electrolysis as well as possible pathways of producing other end-fuels from biogas.

An analysis has been conducted of the integration of renewable energy into the Danish energy system via biogas methanation with electrolytic hydrogen. The

analysis shows that biogas methanation increases the integration of renewable energy. In the reference 2020 model, if all the biogas available in the system is Methanated, by adding 100% buffer capacity and one week of hydrogen storage, electricity produced by offshore wind is increased by 22% in comparison with the constant operation of electrolysis for biogas methanation and no hydrogen storage. Furthermore, methanising all biogas in the system and installing buffer capacity for electrolysis can increase total intermittent electricity share in the energy system by 11%.

In the case of the 2050 Danish energy system model, it is possible to integrate 9% more wind, with biogas methanation (including 100% buffer for electrolysis and one week of hydrogen storage) than in the case of no biogas methanation in the system. The drop in the integration rate from 22% in 2020 model to 9% in 2050 model is due to the already installed electrolysis capacity for the liquid fuel production in the 2050 model. Similar results are in the case of 2035 model.

The additional electrolysis capacity and storage support the integration of renewables, while the results are more sensitive to additional capacity than to the increased storage capacity. Due to the increased electricity consumption, adding biogas methanation to the system increases the electricity system market price. The level of increase depends on the different scenarios and modelled years, but the maximum increase is by 12 €/MWh of electricity in some hours of the year.

Biogas methanation is to play a role in the smart energy system, which requires cross-sectorial connections and electricity storage in the form of heat, gas and liquid fuels. The biogas methanation plants need to be dimensioned with the appropriate hydrogen storage and additional capacity of electrolysis in order to help the renewable energy integration.

BIOGAS RESOURCES

This chapter includes a mapping of the biogas potentials from manure, straw and organic municipal waste in Denmark. The potentials are assessed by a bottom-up approach and summarized at a municipal level.

INTRODUCTION AND APPROACH

The mapping of biogas potentials in Denmark has been carried out in various projects over the years. One of the most recent projects is a report made at the end of 2015 as part of the Danish Energy Agency's biogas travelling team, where SEGES and AgroTech mapped biogas potentials for Denmark [1]. The present report uses a similar approach, but with updated data and focus on the resources that are deemed most useful for power-to-gas production, namely manure, straw and organic municipal waste.

To map resources, a bottom-up approach is used, as data on livestock and crops is quite detailed. The data does not provide a direct overview of the biogas potential; thus, the potentials have to be estimated based on general figures per animal and crop type. For municipal organic waste, the data is not as detailed as data for manure and straw, as it only exists at the municipal level and cannot be disaggregated to smaller spatial points. In Figure 1, the general approach to the mapping is illustrated as an overview, while the following three sections include detailed explanations of the methods, followed by a chapter with maps of each resource at the municipal level.

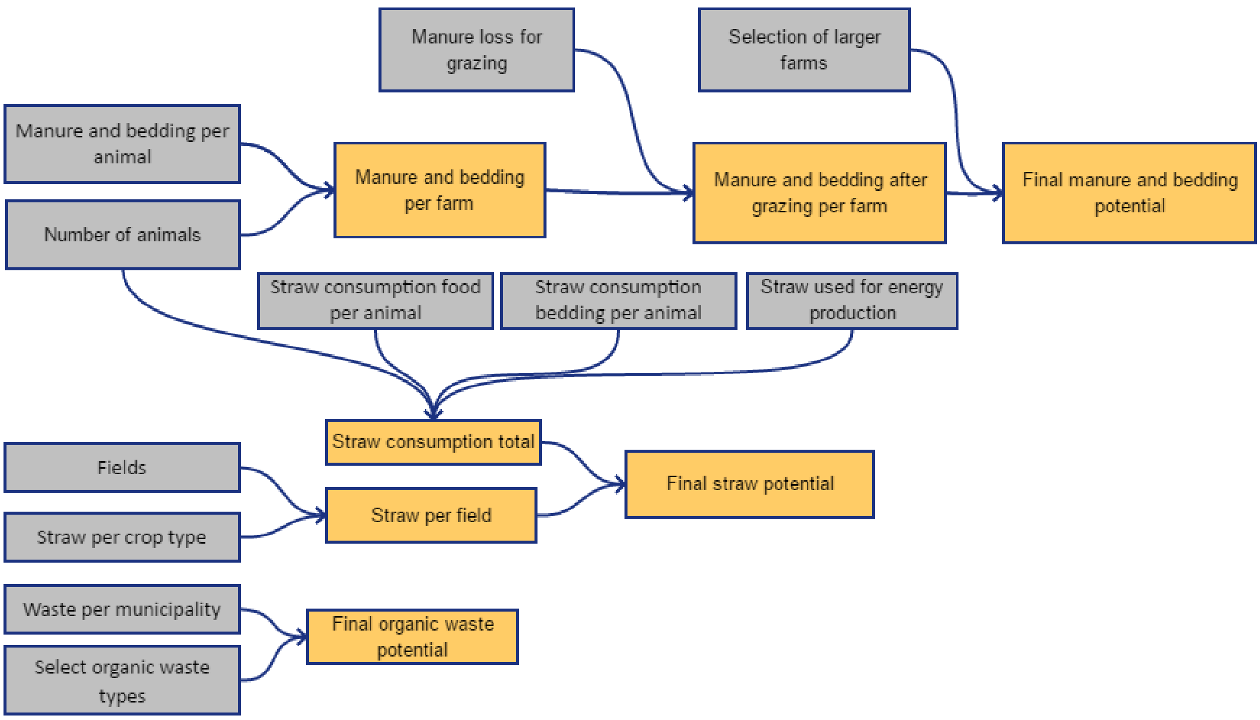

Figure 1: General methodology

MAPPING OF MANURE AND BEDDING

As mentioned in the introduction, the first part of the analysis is the mapping of manure and bedding from animal livestock. As the data in Denmark is quite comprehensive within the agricultural sector with 35 types of animals and 35 types of use, the focus in this report will be limited to two types of animals, cattle and pigs, and only the animals used for meat and dairy production. Based on [2], these two types of animals provide approximately 90% of the usable manure and bedding. Thus, it is assumed that another 10% could be gained from other types of animals. In Table 1, the manure and bedding production per animal for cattle and pigs is presented and divided depending on the end use and the age.

Table 1: Manure and bedding per animal in ton per year. Based on [3] *for cows and* [4] *for pigs.*

Number		Animal	Manure	Bedding
1	Cows	Cattle (meat production)	10.18	11.31
2		Dairy cows	25.81	-
3		Other cows	6.67	4.85
4		Calves	3.31	2.10
5	Pigs	Sows	4.00	-
6		Other pigs	1.60	-
7		Piglets	0.20	-

In this report, the potential is estimated based on data from The Central Livestock Register 2018 database [5]. This register is a comprehensive database that for 2018 included 36,436 address level farms. To assess the biogas potential, the initial step was to select only cattle and pigs from the database. After this, the codes from Table 1 were added to each farm and the content of the table was joined to each farm. As the database includes information on the number of animals on each farm, the total potential was estimated by multiplying the manure and bedding per animal with the total number of animals. Part of the potential cannot be used for biogas production, as it is lost during the grazing of the animals. For conventional cattle, this loss is assumed to be 13%, for organic cattle 22% and for organic pigs 14%. Thus, these shares were subtracted from the total potential on each farm, based on the type of use. Finally, due to economies of scale, only the larger farms were chosen. In this report, larger farms are determined to be farms with more than 750 tons of manure or 300 tons of bedding per year.

With these assumptions, the total amount is 27,632,435 tons of manure and 2,315,437 tons of bedding (see Table 2). In [2] the potentials were found to be 32,446,000 tons of manure and 2,811,000 tons of bedding for 2012-2013. The estimate in this report is slightly lower, which can be attributed to differences in both year and methodology. As manure and bedding have different components, the dry matter content for each is estimated. For manure, a dry matter content of 7.9%

for cattle and 5.8% for pigs is used, while for bedding, 25% is used for cattle. This gives totals of 1,873,527 tons of dry matter for manure and 578,859 tons of dry matter for bedding, which means that bedding is around 30% of the total potential.

Table 2. Potentials for manure and bedding both in Denmark divided by the animal type

Animal	Manure (ton)	Bedding (ton)	Manure (dry ton)	Bedding (dry ton)
Cattle	12,897,453	2,315,437	1,018,899	578,859
Pigs	14,734,982	-	854,629	-
Sum	27,632,435	2,315,437	1,873,528	578,859

MAPPING OF STRAW

The mapping of straw resources is based on two steps; first estimating the total straw production, followed by an estimate of the straw used for food and bedding for animals as well as heat and electricity production.

For the mapping of the total straw production, the main dataset used is the Danish Field Database [6]. This database provides data on the hectares of land as well as the crop type for each field. As the database does not provide direct information of the straw quantities, these have to be estimated based on the type of crops and the soil type for the land area. Thus, the Danish Soil Classification Map [7] with 9 soil types is combined with the field database to give a dataset that provides crop type, soil type and the area for each field. The Danish Field Database is also comprehensive with 304 types of crops. To simplify the calculations, the estimate is only based on the 8 most common types. Table 3 presents the estimates on straw as tons of dry matter per hectare for each of the 8 crop types and 9 soil types.

Table 3: Straw production for the most important crops divided into soil types, straw per ton of dry matter per hectare [2].

Crop Code	Crop Text	Soil type 1 and 3	Soil type 2 and 4	Soil type 5-6	Soil type 7-9
1	Spring barley	2.0	2.1	2.7	2.9
10	Winter barley	2.4	2.4	3.3	3.6
11	Winter wheat	2.7	3.0	3.8	4.1
14	Winter rye	3.9	4.5	5.5	5.9
16	Triticale	3.9	4.4	5.1	5.4
22	Winter rape	2.5	2.9	3.3	3.5
30	Peas	3.0	3.0	3.0	3.0
252	Seed grass	3.0	3.0	3.0	3.0

When combining the Danish Field Database with the estimates from the table, the total straw potential is estimated at 3,169,622 dry tons, or with a dry matter percentage of 85%, a total of 3,728,967 tons. Table 4 summarizes the results for different types of straw.

Table 4. Straw potential by different types mapped

Crop Code	Crop text	Dry ton	Ton
1	Spring barley	1,509,928	1,776,386
10	Winter barley	186,734	219,687
11	Winter wheat	847,160	996,659
14	Winter rye	45,385	53,394
16	Triticale	12,725	14,970
22	Winter rape	314,424	369,911
30	Peas	15,462	18,190
252	Seed grass	237,806	279,772
	Sum	**3,169,623**	**3,728,968**

This is lower than the estimate of 5,589,000 tons from the Danish statistics for 2014 and from the 5,234,000 tons mapped in the [2]. This difference is due to deviations in model year and methods, where the output from [2] includes more crop types. The mapping conducted in this report lacks data due to data unavailability from public sources.

As mentioned in the approach, the existing straw consumption needs to be subtracted from the straw production to estimate the potential for biogas production. The first demand is the straw consumption for animals, which in this case is only for the cattle, corresponding to 700 kg/year for old animals, 250 kg/year for younger animals, and 150 kg/year for calves. Bedding is assumed to be 62% of the amount needed for food, which gives a total straw consumption of 576,621 tons/year for food and 357,505 tons/year for bedding. In total, 934,126 tons/year is straw for animals. It should be mentioned that large uncertainties relate to the assumptions behind this assessment.

Another large straw consumption is used for energy production. To assess the geographic distribution of straw demand for energy consumption, the Danish Energy Agency's Energy Producer Statistics from 2015 are used [8]. In total, the demand for energy consumption corresponds to 2,004,504 tons/year in 2014 with an energy content of 14.5 GJ/ton of straw.

MAPPING OF BIODEGRADABLE MUNICIPAL WASTE

The data input for biodegradable/organic municipal waste is based on data from 2016 from the Waste Statistics made by the Danish Environmental Protection Agency. The input data concerns the municipality level. To select the organic waste, only 6 types are chosen based on the categories in The European Waste Classification and a six digit code[1] in the brackets.

a. Grease and oil mixture from oil/water separation containing only edible oil and fats (19 08 09)
b. Biodegradable kitchen and canteen waste (20 01 08)
c. Edible oil and fat (20 01 25)
d. Biodegradable waste (20 02 01)
e. Mixed municipal waste (20 03 01)
f. Municipal waste not otherwise specified (20 03 99)

For *a-d*, it is assumed that everything is biodegradable, but for *e* and *f* only 55% is assumed to be biodegradable. This gives 2,960,387 tons of organic waste in total (see Table 5 for more details).

Table 5. Biodegradable waste potential divided into categories

EWC classification	1000 tons
19 08 09	6.412
20 01 08	231.85
20 01 25	3.43
20 02 01	1,153.16
20 03 01	1,488.36
20 03 99	77.17
SUM	**2,960.39**

According to [9], the amount of waste from these categories is approximately the same over the previous years, i.e. around 2.9 million tons of waste including dry matter. Category b, biodegradable kitchen and canteen waste, has been the highest growing category, due to new regulations.

MUNICIPAL DISTRIBUTION OF BIOGAS SOURCES

This section presents the results of the mapping, showing the spatial distribution of the three categories of biogas resources. The first category is manure and bedding from animals, which is illustrated in Figure 2. From the maps, it is clear that both manure and bedding are more dominant in the western part of the country. As the values are given in tons and not dry tons, the potentials for manure look much larger than for bedding. However, in dry tons the values would be more similar and bedding would correspond to 30% of the manure potential.

[1] The different types of waste in the European Waste Classification are fully defined by the six-digit code for the waste

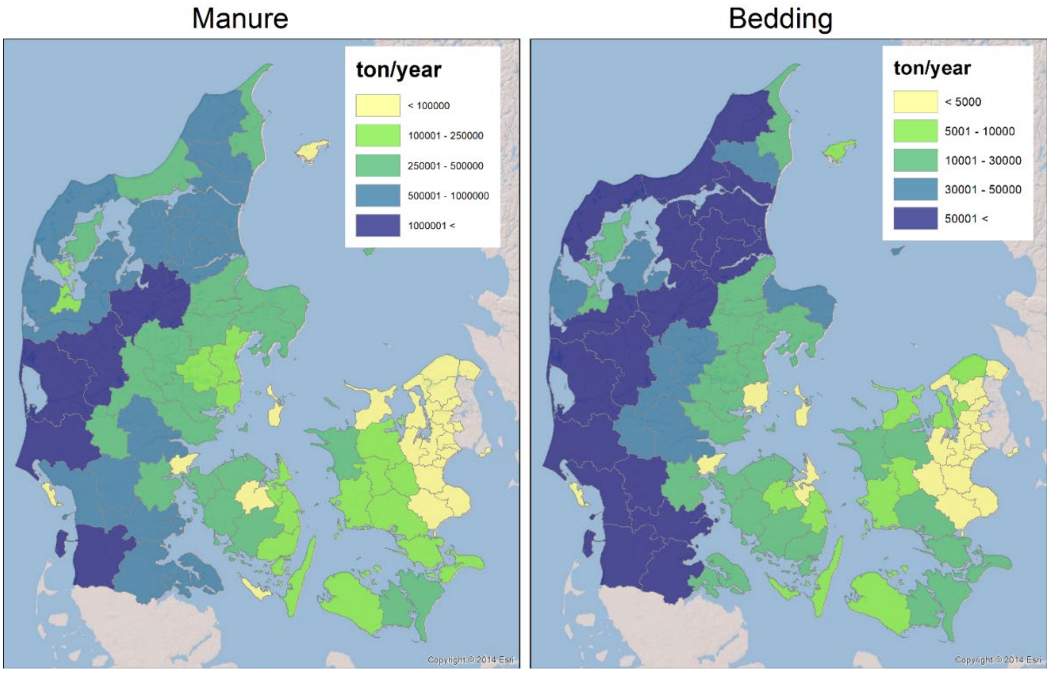

Figure 2: Manure and bedding potential on the municipal level

The next category is the potential from straw, as shown in Figure 3. The figure shows both the straw demand and the straw production. The demand includes the demand for animal feed, bedding and energy production. It is clear that there is a straw demand mainly in the western part of the country and in the larger cities, which is caused by the demand for heat and electricity production. The straw production, on the other hand, takes place outside the larger cities. It seems to be more spread across the country, with a larger production in the eastern part as well.

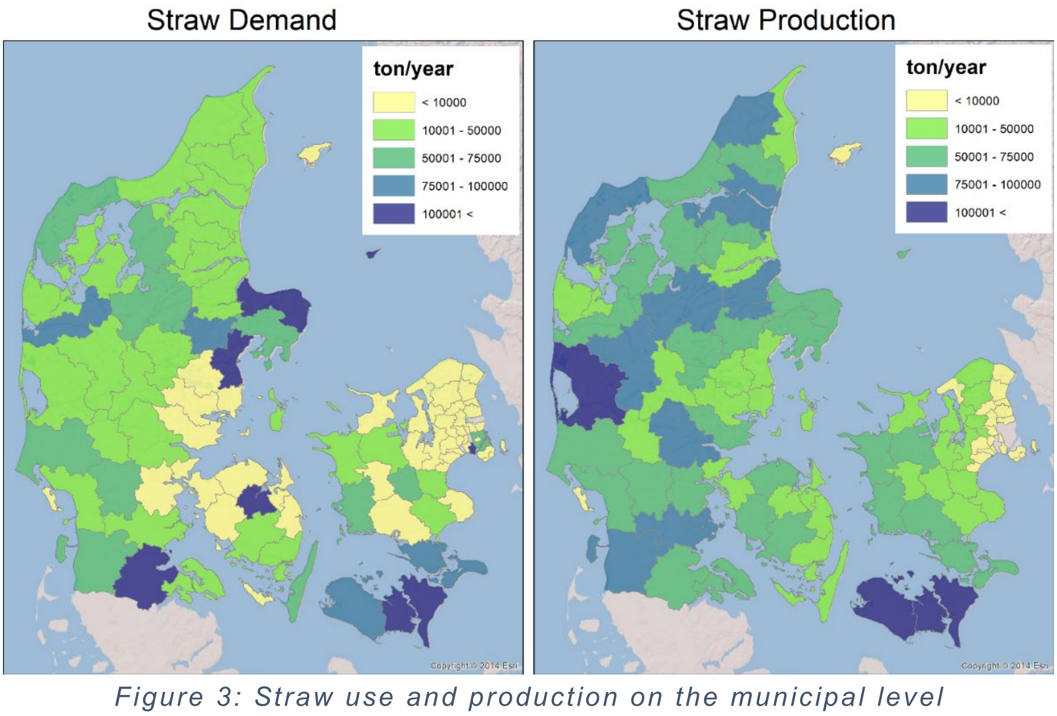

Figure 3: Straw use and production on the municipal level

The third category is organic/biodegradable waste, which is shown in Figure 4. Here the potential is around the larger cities and in the larger municipalities.

Organic Waste

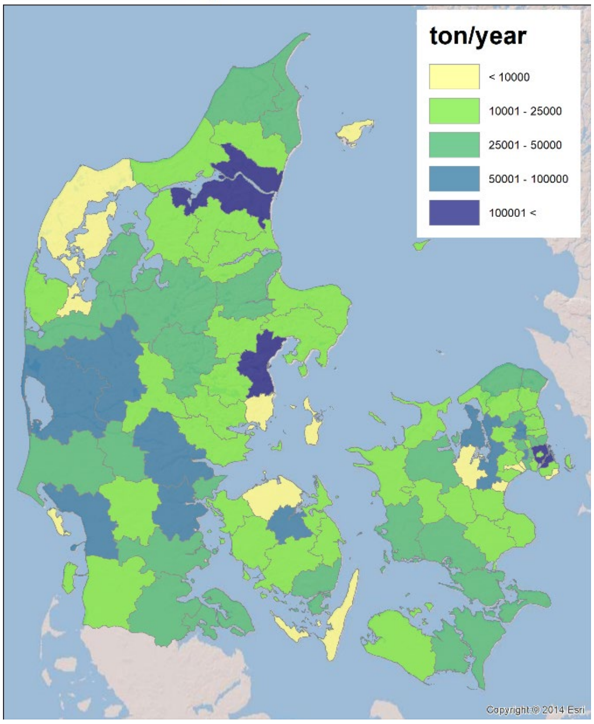

Figure 4 Organic Waste potential on the municipal level

Summary of the results and the biogas potential based on the mapped resources is presented in Table 6. As the results are very sensitive to the methodology and the energy properties of the resources results can vary from the other reported potentials. Potential mapped in this report is lower than the potential reported in [2].

Table 6.Biogas resources potential based on the mapping output.

	Manure / Bedding	**Straw**	**Organic waste**	**Total**
Biomass potential [tons]	27,632,435 / 2,315,437	3,728,967	2,960,387	36,637,226
Of which dry matter [tons]	1,873,527 / 578,859	3,169,622		5,622,008

According to [2], in 2015, 91 biogas plants have used 11.9 mio tons of biomass per year where 2.2 mio tons were included for 16 plant that were in the planning phase. In the 2020 projection, 18.5 mio tons of manure is used for biogas production, which represents 50% of the total manure, fulfilling a national goal of 50% utilisation of manure for energy purposes [10].

BIOGAS PRODUCTION

Biogas can be produced from various biodegradable materials such as organic waste, animal manure or energy crops. The focus here will be on agricultural residues and organic waste. In an anaerobic digestion process, microorganisms ferment the organic material from the wet biomass into a mixture of methane and carbon dioxide. This process takes place in the absence of oxygen [11]. In order to secure the optimal digestion, the temperature in the reaction tank is heated to either 35-40°C (mesophilic digestion) or 50-55°C (thermophilic digestion) [12]. According to Neshat et al. [13], thermophilic digestion can improve the performance of the anaerobic digestion as the solubility of the organic compounds as well as the chemical and biochemical reaction rates are higher. However, thermophilic digestion requires more energy to heat up the reactor and the mesophilic digestion can enhance the process stability and pathogen inactivation. The material is further processed in a post-digestion tank to produce more gas [12].

The hydraulic retention time (HRT) should be carefully determined as an extended HRT can kill the microorganisms due to the lack of nutrients, while a limited HRT can result in cell intoxication or low methane yield [14]. In Denmark, the HRT is normally less than 25 days. Danish biogas plants use continuous digestion in fully stirred digesters. This is done by removing an amount of digested biomass, which is replaced by a corresponding amount of undigested biomass. This procedure is typically done several times a day. Residues from the reaction tank are stored and become digested along with the residue from the post-digestion tank. This digestate is one of the outputs which is a valuable fertilizer due to the content of nutrients [12].

The digestate can also be used in air gasifiers to produce additional gas and the by-products, biochar and ashes, of the gasification process can then be used as fertilizers [15]. Using co-substrate in the process can improve the quality of the digestate as more nutrients are preserved, which can make the biogas production more economically viable [16]. Typically, raw biogas has a methane (CH_4) content between 50 and 70% and a carbon dioxide (CO_2) content of 30-50%. Additionally, also a minor share consists of hydrogen (H_2), nitrogen (N), oxygen (O), hydrogen sulphide (H_2S) and ammonia (NH_3) [12,17,18]. The content of volatile solid in the biomass has a significant influence on the output, as this represents the part of the biomass that may be converted into biogas. The input of digestible material represents different volatile solid contents. The volatile solid content is approximately 75% for animal slurry and around 80% for separated household waste [12]. Table 7 below shows the energy content of various biomass inputs.

Table 7. Energy properties based on biomass inputs from a basic biogas plant and increased industrial organic waste input and increased straw input. Source (DEA, 2019)

Basic biogas plant	Methane production (GJ/ton)	Input share: Basic mix (% of mass input in tons)	Methane production: Basic mix (% of total energy)	Input share: 5% Industrial organic waste (% of mass input in tons)	Methane production: Industrial organic waste (% of total energy)	Input share: Increased straw (% of mass input in tons)	Methane production: Increased straw (% of total energy)
Pig & cattle slurry	0.44	79.8%	44%	75.8%	34%	73.3%	26%
Deep litter	2.00	8.0%	20%	8.0%	16%	8.0%	13%
Manure, stable	1.57	6.1%	12%	6.1%	10%	6.1%	8%
Straw	7.27	0.0%	0%	0.0%	0%	6.3%	37%
Industrial organic waste	4.83	1.0%	6%	5.0%	25%	1.0%	4%
Household waste	3.41	1.6%	7%	1.6%	6%	1.6%	4%
Energy crops	1.5-3.5	0.0%	0%	0.0%	0%	0.0%	0%
Other	1-5	3.5%	11%	3.5%	9%	3.5%	7%
Total (GJ/ton)	-	0.8	100%	0.97	100%	1.20	100%

As Table 8 indicates, straw and industrial organic waste are the biomass inputs with the highest energy content. The input mix for a basic biogas plant allows a total methane production of 0.8 GJ/ton. An increase in the industrial organic waste will lead to a total methane production of 0.97 GJ/ton. When increasing the input of straw, the total methane production will increase to 1.20 GJ/ton. The increase in the methane output is mainly corresponding to the lower amount of water in the biomass mix. Increasing the input of deep litter and straw requires a special plant design with a pre-treatment of the feedstock. The DEA assumes an upper limit of straw and deep litter material of 50% of the methane production. The increase of industrial organic waste requires more transport concerning the supply of biomass [12]. In the table below, the energy content of relevant biomass types and their respective costs are shown.

Table 8. Energy content for relevant biomasses and costs. Source [12]

	GJ/ton	Price per ton (€) incl. transport
Pig & cattle slurry	0.44	3.36
Straw	7.27	67.4
Industrial organic waste	4.83	40.3

As shown in Table 7, the biomass types with higher methane yield are more expensive to transport and this will have an impact on the operation and maintenance (O&M). Thereby, an increase in the yield of methane will also increase the costs. In Table 9, financial data is listed based on a basic biomass input presented in the table. In another report from the DEA, the costs for Danish biogas plants have been collected and presented. The Biogas Taskforce project identified the production costs of six biogas plants, which were between 11 and 23 €/GJ. Three plants described in DEA's technology catalogue have costs of 14-17€/GJ and ten other plants in the range of 16-21€/GJ [19].

Table 9. Data sheet for biogas plant with basic configurations. Adapted from [12]

Technology	**Biogas plant, basic configuration**			
	2015	**2020**	**2030**	**2050**
Input				
Biomass (tons/year)	356,000	356,000	356,000	356,000
Aux. electricity (kWh/ton input	3.7	3.8	3.8	3.8
Aux. process heat (kWh/ton input	18.6	18.6	18.6	18.6
Output				
Biogas (GJ/ton input)	0.80	0.75	0.75	0.75
Lifetime	20	20	20	20
Financial data				
Specific investment (M€/MW output)	1.81	1.71	1.54	1.39
Total O&M (€/MW/year)	198,785	194,715	197,702	195,722
Total O&M (€/ton input/year)	5.03	4.63	4.70	4.66
Methane emissions (Nm^3 CH_4/ton input/year)	0.44	0.42	0.42	0.42

The DEA has made projections until 2050 showing the expected price reductions. Table 10 shows financial data concerning additional costs when increasing the industrial organic waste or the input of straw. The different inputs of straw and organic waste have an equal energy output when converted, which makes the costs comparable. The operation and maintenance costs are lower when handling additional straw until 2020. From 2030, the O&M costs are lower concerning industrial organic waste. The investment cost for handling straw in the feedstock mix is significantly higher than facilitating an additional input of organic waste [12].

Table 10. Data sheet for additional industrial organic waste and additional straw in the feedstock mix. Source (The Danish Energy Agency, 2019)

Technology	**Biogas plant, additional industrial organic waste in the feedstock mix**				**Biogas plant, additional straw in the feedstock mix**			
	2015	**2020**	**2030**	**2050**	**2015**	**2020**	**2030**	**2050**
Input								
Additional input (tons/year)	6,529	6,529	6,529	6,529	4,337	3,957	3,957	3,957
Aux. electricity (kWh/ton additional input	10.30	10.30	10.30	10.30	63.00	63.00	63.00	63.00
Aux. process heat (kWh/ton additional input)	18.60	18.60	18.60	18.60	18.60	18.60	18.60	18.60
Output								
Biogas (GJ/ton additional input)	4.8	4.8	4.8	4.8	7.3	8.0	8.0	8.0
Lifetime	20	20	20	20	20	20	20	20
Financial data								
Investment (€/MW output)	276,050	276,050	276,050	276,050	407,676	371,930	371,930	371,930
Investment (€/ton additional input/year)	42.28	42.28	42.28	42.28	94.00	94.00	94.00	94.00
Total O&M (€/MW/year)	49,500	49,904	52,056	53,132	47,387	44,727	52,704	56,692
Total O&M (€/ton additional input/year)	7.6	7.6	8.0	8.1	10.9	11.3	13.3	14.3
Methane emissions (Nm^3 CH_4/ton input/year)	4.0	4.4	4.4	4.4	4.0	4.4	4.4	4.4

Biogas can be used directly for electricity and heat production either in CHPs or boilers for process heat and space heating. The biogas can be further purified or methanated to methane by using different technologies. There are some major advantages of upgrading biogas as it reduces greenhouse gas emissions (GHG) and emits less hydrocarbon, nitrogen oxide and carbon monoxide in comparison with conventional gasoline or diesel [20].

BIOGAS PRODUCTION STATUS IN DENMARK AND FUTURE POTENTIAL

Denmark is a country rich in biomass resources and is one of the regions in Europe with the highest biomass residual potentials [21]. However, in 2017, the import of different biomass in Denmark has reached 42% of the total biomass consumption (see Figure 5). Therefore, the utilisation of local resources and self-sustainability are important focus areas in the transition towards future energy systems.

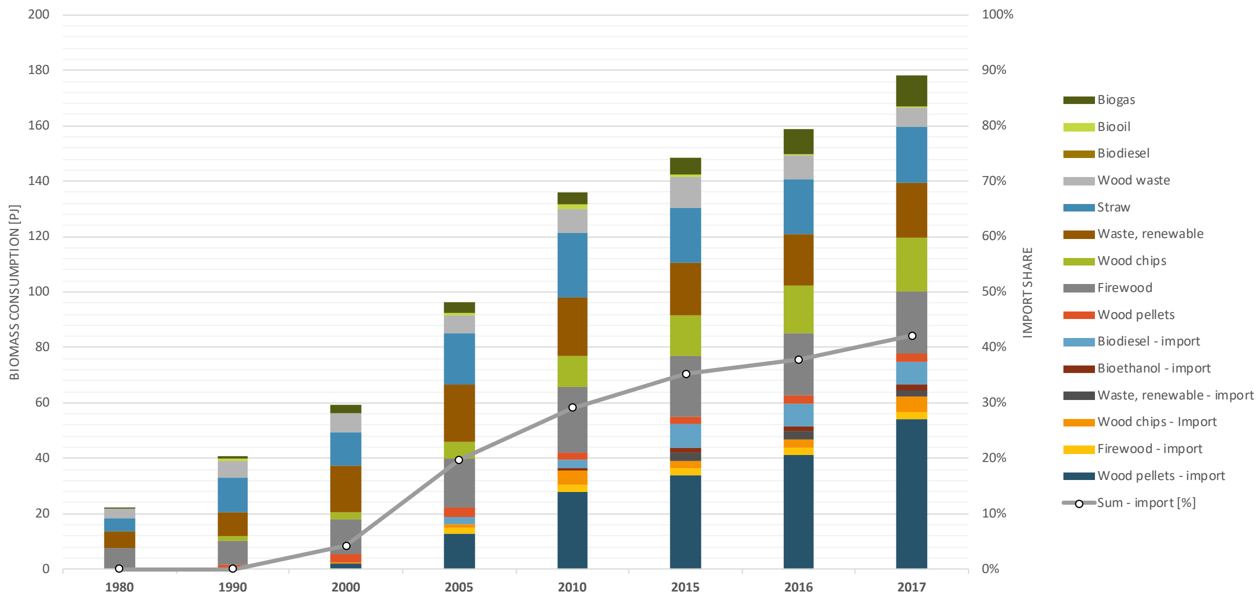

Figure 5. Biomass consumption including import in Denmark from 1980 to 2017

Biogas production has a long tradition and is a renewable alternative to fossil natural gas. In 2017, biogas production represented 6.5% of the renewable energy production in Denmark. The production has increased from 0.2 PJ in 1980 to 11.16 PJ in 2017 (see Figure 6). From 2015 to 2017, the biogas production has increased by 44% [22] and it has been further increased in 2018. The majority of the biogas used in Denmark is used directly without purification (CO_2 removal). The use of biogas for power production and the upgrade of biogas to grid quality, industrial processes, transport, and heat production are supported by the Danish government. However, currently there is no support for biogas methanation with the addition of hydrogen. In 2018, as a part of the new Energy Agreement, an annual amount of 240 million DKK was dedicated to the expansion of biogas and other green gases over the next 20 years [23].

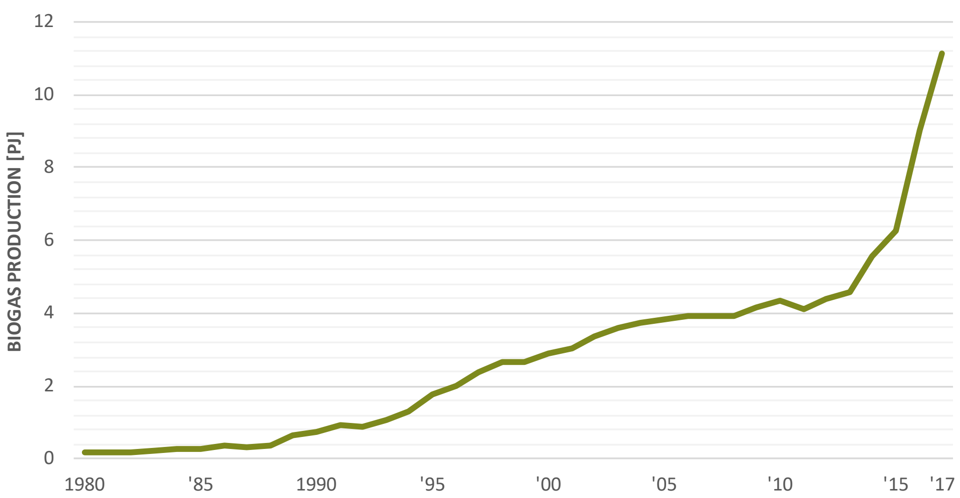

Figure 6. Biogas production in Denmark from the 1980s to 2017

In Denmark, there are currently 163 biogas plants, of which 50% are based on agriculture, 31% are sewage treatment, 3% are based on industries and 16% on landfills (see Figure 7). As of 2016, 47% of the biogas was used for electricity and DH production, while the rest was delivered to the gas grid, used in industry and transport [22]. In 2016, Denmark had 18 biogas purification plants supplying the natural gas grid with biomethane. The first full-scale biogas upgrading plant based on wastewater treatment was established in Fredericia in 2011 [24]. Today, biogas is delivered to the gas network from 33 biogas plants [25].

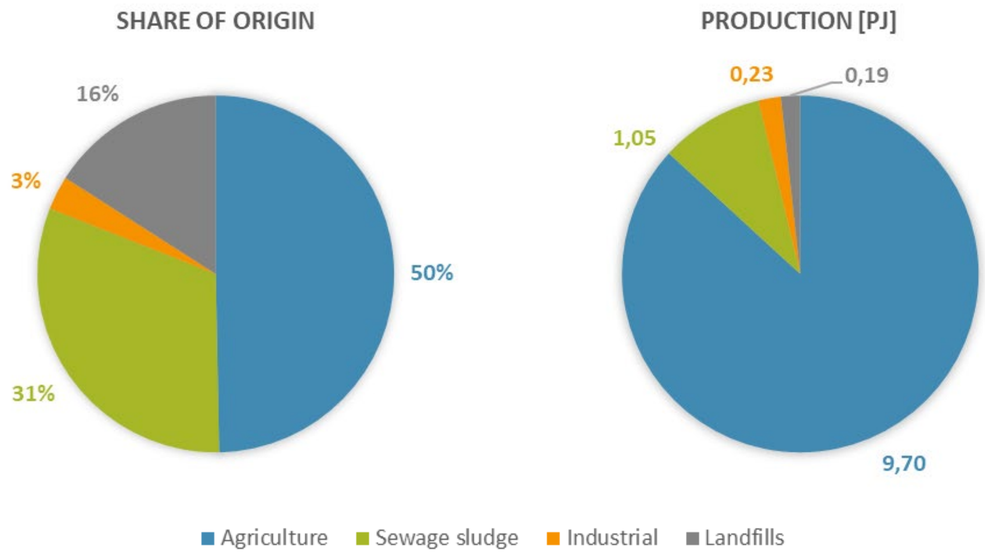

Figure 7. Share of different types of biogas plants in Denmark and biogas production as of March 2017 [26]

In 2013, Denmark was a moderate biogas market at the EU level, based on six biogas upgrading installations according to [27]. However, the number of biogas upgrading plants in Denmark has increased significantly in the last 6 years, making Denmark a mature market for this technology.

According to Gylling et al. [28], additional 10 million tons of biomass can be produced in Denmark by 2020, compared to the biomass production in 2009. The potential biomass production is based on three scenarios, a business-as-usual (BAU) scenario, a biomass-optimised scenario and an environment-optimised scenario, where both agriculture and forestry are adjusted to produce the maximum level of biomass. This additional biomass potential covers a wide range of biomass types, also including biomass which is not suitable for biogas production. The largest potential is found in green biomass, like grass and beet, followed by manure and straw, which are all suitable for biogas production.

Based on the additional biomass potential from [28], Møller and Jørgensen [29] have presented three biogas technology scenarios and related methane potential. Figure 8 illustrates the methane potential for 2035 where three scenarios: state-of-the-art, optimised technology and optimised technology + methanation are included.

The biogas methanation in the figure presents the potential for e-methane produced from biogas methanation with electrolytic hydrogen.

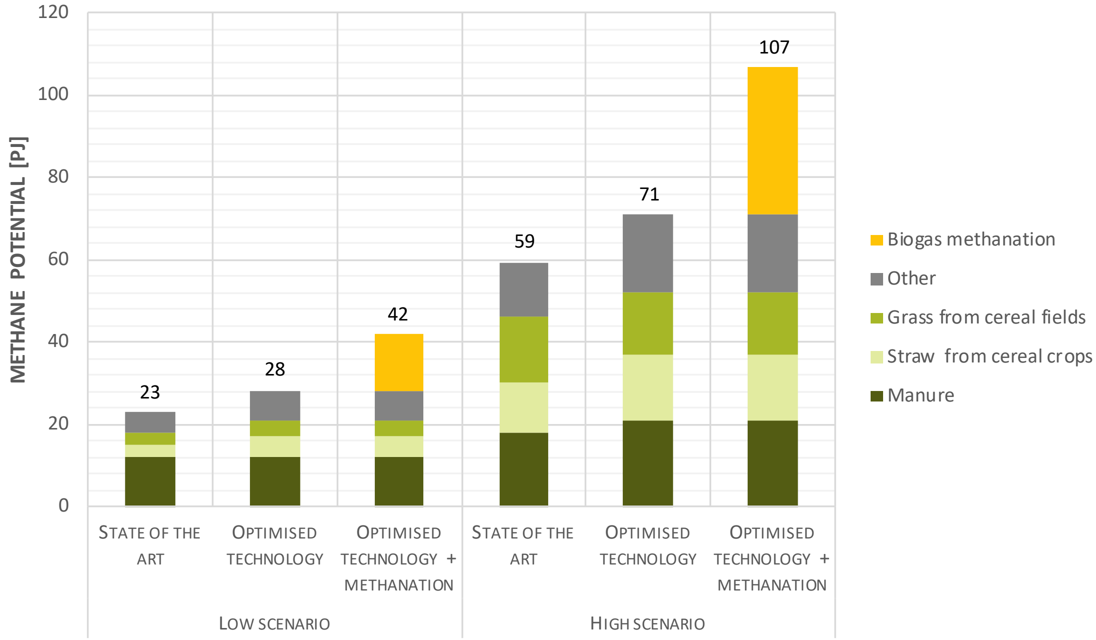

Figure 8. Biomethane and electro-methane potential for Denmark based on different scenarios. Adapted from [30] *and* [31]

As different projections can be seen of the biogas potential including additional biogas methanation, different sources have been reviewed and illustrated in Figure 9. The biogas potential without additional biogas methanation is shown in dark green and biogas methanation potential in light green. The results range from 32 PJ to 107 PJ of the total methane that can be produced, if we methanise the CO_2 part of biogas with electrolytic hydrogen. This wide range of the potentials indicate that different methodological assumptions can lead to different results and this report continues using the lower range of the potentials that were used in energy modelling by [32,35,36].

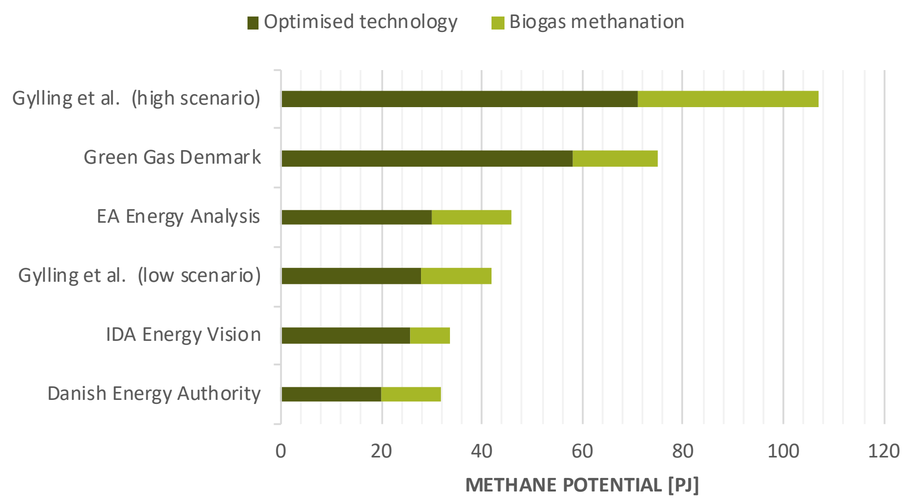

Figure 9. Biogas (biomethane and electro-methane) potential for 2035 according to different sources [30,32–35]

STATE-OF-THE-ART OF BIOGAS PURIFICATION AND BIOGAS METHANATION

The upgrading methods can be divided into two categories:

- removal of the CO_2 fraction from the biogas, and
- methanation of biogas, where the addition of hydrogen from another source reacts with the CO_2 content in the biogas [17].

These two methods will be described in detail below including the state-of-the-art of electrolysis used for producing the hydrogen needed for biogas methanation.

BIOGAS UPGRADING (PURIFICATION) BY CO_2 REMOVAL

In the biogas upgrading and cleaning, the main purpose is to remove the CO_2 content in order to meet the quality specifications for natural gas in the grid. Likewise, it is also necessary to remove particles, water moisture, ammonia, hydrogen sulphide and nitrogen depending on the composition of the raw biogas [37,38]. However, nitrogen is rarely removed as it is an expensive procedure [12]. Hydrogen sulphide is mainly targeted to be removed as it is corrosive gas [37,39]. Biogas upgraded to biomethane can be injected into the natural gas grid where it can be stored; it can be compressed and stored outside of the grid or it can be used as a renewable fuel for transport.

There are six available upgrading technologies today, not all of them are equally commercially mature, and two R&D technologies*:

- Water scrubbing
- Chemical absorption (amine scrubbing)
- Pressure swing adsorption (PSA)
- Membrane separation
- Organic physical scrubbing
- Cryogenic*
- Enzymatic*

Water scrubbing is the most commonly used upgrading technology [37,38]. The absorption process in the water scrubbing technology is purely physical. Water is used to wash out the content of both CO_2 and hydrogen sulphide as these gases are more soluble in water than methane [37]. There is no need for further compression of the methane to the natural gas grid, as the pressure in the water scrubber is typically higher than the pressure in the natural gas distribution grid [12]. An advantage of the scrubber is that it is non-corrosive [39].

Amine scrubbing has the highest efficiency in the conversion of methane and uses chemical absorption of CO_2. The scrubbing technology can be integrated using the excess heat from other high-temperature (120-150°C) processes. However, it is unlikely to find a waste heat source at the plant site with this temperature range. The excess heat of around 65°C from the amine scrubber itself can be used in other low-temperature applications, e.g. a biogas digester. The amine scrubber needs electricity as an input for compression of the gas for grid injection [12]. One drawback of the amine scrubber can be, in contrast to the water scrubber, that it uses corrosive absorbents [39].

The PSA scrubbing technology separates some gas components from a mixture of gases under high pressure in accordance with the component's molecular characteristics and affinity for an absorbent material, which is often active carbon. To desorb the absorbent material, the process then swings to low pressure [12,37]. The vast majority of the PSA scrubbing technology is located in Sweden and Germany, while there is currently no such plant operating in Denmark [12].

The membrane separation technology consists of bonded hollow fibres that are permeable to ammonia, carbon dioxide and water. Both hydrogen and oxygen flow through the membrane to some extent, while methane and nitrogen only flow through to a very low extent. This process is typically carried out in two stages. Before meeting the membranes, water and oil droplets from the gas are first caught in a filter. Active coal is hereafter often used to remove hydrogen sulphide from the gas [12]. The organic physical scrubbing technology functions in the same way as the water scrubber, but the CO_2 is here absorbed in an organic solvent instead of water [37].

The cryogenic upgrading technology is an additional path for upgrading biogas into biomethane. This technology can produce liquified biomethane (LBG) and remove nitrogen from the biogas. Cryogenic upgrading may offer a lower energy demand than the abovementioned upgrading technologies [12]. However, the technology deployment has been limited due to operational problems and is still in the research and development state [37,38].

The enzymatic upgrading technology is a new technology under development that potentially provides a route, which in comparison with the commercially available upgrading technologies is both more energy-efficient and can reduce the production costs of biogas upgrading by 25%. Additionally, it is expected that the new upgrading technologies will reduce the energy consumption by around 50% [12].

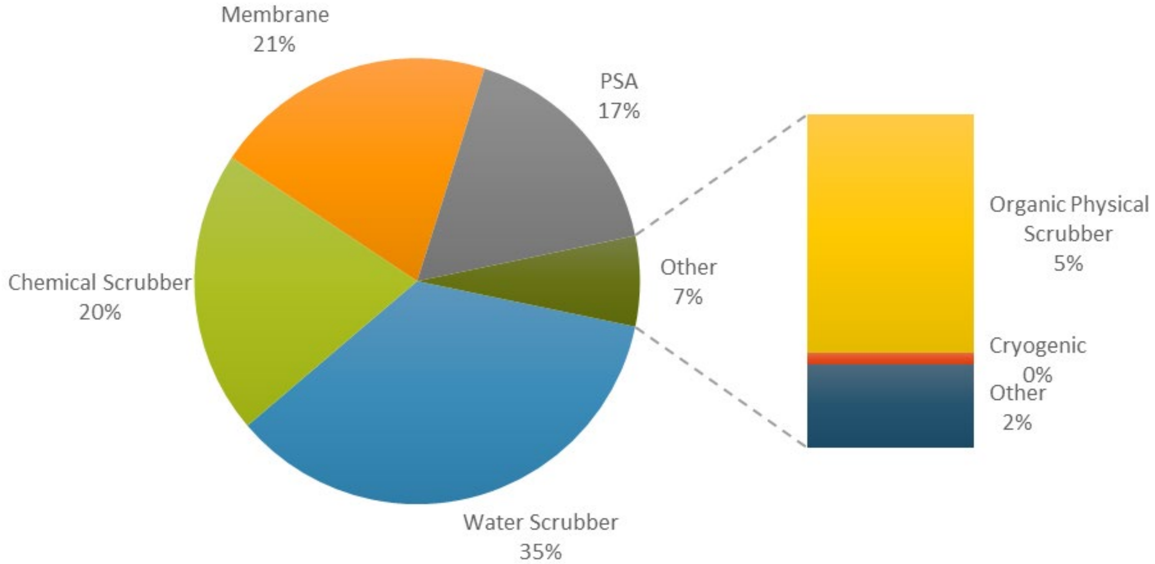

Figure 10. Share of upgrading biogas plant technologies on the global scale for 2015 (adapted from [37]*)*

In 2015, 428 plants globally have a distribution of commercial technologies, as illustrated in Figure 10. The typical upgrading technology varies in capacity dependent on the specific type of upgrading technology and the location. The typical size for newer plants in Denmark is between 1,000 and 2,000 Nm^3 per hour of biomethane. In Germany, most biogas upgrading plants have a capacity between 700 and 1,400 Nm^3 per hour of raw biogas, while the most common plants in Sweden produce about 600, 900 and 1,800 Nm^3 raw biogas per hour [12]. Table 11 below presents the data and projection for a biogas upgrading plant.

The production of biogas and biogas upgrade result in fugitive emissions / methane slippage. The literature reports fugitive emissions that vary between 1 and 7% of the produced biomethane [40]. Methane emissions from the existing biogas upgrading plants show methane losses between 0.07% and 1.97% [41]. While amine based upgrading technologies have the lowest methane losses, the water scrubber has the highest leakages. It is assumed that one of the newer developed upgrading technologies will take over from 2030 and that the slip of methane from these technologies will be close to zero.

Table 11. Biogas upgrading technology data. Source (The Danish Energy Agency, 2019)

Technology	Biogas upgrading			
	2015	2020	2030	2050
Energy data				
Typical plant size (MJ output)	5.92	5.92	5.92	5.92
Typical plant size (Nm^3 biogas/h)	1,000	1,000	1,000	1,000
Capacity (Nm^3 biomethane/h)	594	594	594	594
Inputs				
Biogas (% of biogas input)	100	100	100	100
Auxiliary electricity for upgrading (% of biogas input)	4.3	4.3	2.2	2.2
Auxiliary electricity for compression (% of biogas input)	1.0	1.0	1.0	1.0
Output				
Biomethane (% of biogas input)	99	99	100	100
Waste gas (% of biogas input)	1	1	0.1	0.1
Waste heat (% of biogas input)	5.3	5.3	3.2	3.2
Technical lifetime (years)	15	15	15	15
Financial data				
Specific investment, upgrading and methane reduction (€/MJ/input)	335,000	302,000 (268,000-318,000)	272,000	245,000 (172,000-287,000)
Specific investment, grid connection at 40bar (€/MJ/input)	134,000	121,000 (107,000-127,000)	109,000	98,000 (69,000-115,000)
Fixed O&M (€/MJ/input/year)	11,800	10,600 (9,400-11,200)	9,500	8,600 (6,000-10,100)
Variable O&M (€/GJ/input)	0.93	1.03	0.88	1.02
Technical specific data				
Methane slip (%)	1	1	0.1	0.1
Minimum load (% of full load)	50			
CO_2 removal (%)	98.5			

**Figures in parenthesis presents uncertainties associated with the specific projections.*

ELECTRO-METHANE BY HYDROGEN ADDITION

Carbon dioxide from the production of biogas can be utilised to produce electro-methane by adding hydrogen (H_2) from electrolysis to the biogas produced via anaerobic digestion. This can be an effective way of storing excess electricity from an intermittent renewable energy source (RES), as the conversion allows fluctuating energy to be stored as a chemical energy [17,39,42]. The method is also called power-to-gas (P2G) and can be used to store surplus electricity in the form of a gas by using the large storage capacity of the natural gas grid. Simultaneously, the addition of hydrogen to the biogas is a more efficient way of utilising the biomass resources, as the carbon dioxide is used in the production of electro-methane and not discarded as a waste product, as it is in the conventional upgrading of biogas [17].

A review of the electrolysis and biogas methanation technologies including the regulation abilities of these is presented below.

ELECTROLYTIC HYDROGEN

The carbon dioxide fraction in the biogas can be utilised through methanation by adding hydrogen to the process. Pure hydrogen from a renewable energy source can be obtained from an electrolysis technology. Electrolysers use electricity to split water into hydrogen and oxygen between two separated electrodes. The three main electrolysers available are alkaline electrolysis (AEC), proton exchange membrane electrolysis (PEM) and solid oxide electrolysis (SOEC) [17,43]. Alkaline is the most mature electrolysis technology and has been used in the industry for more than a century. PEM is also a commercially available electrolyser and, as it has the ability to operate in a more flexible energy system, it is rapidly getting more widespread on the market. SOEC is still in the development phase but the electrolyser contains a large potential in comparison with both AEC and PEM, due to its high energy efficiency and expected lower future costs [17,43,44].

Both AEC and PEM electrolysis are classified as low-temperature electrolysers as their operating temperature is below 100°C, while SOEC is high-temperature with operating temperatures up to 1000°C [39]. According to Brynolf et al. [43], alkaline electrolysis typically operates at a temperature in the range of 60 to 80°C and either under an atmospheric or pressurised condition with an efficiency between 43 and 69%. The typical operation temperature of a PEM electrolyser is about 50 to 80°C and it has the ability to operate under a higher pressure than AEC electrolysers, i.e. around 80 bar or more. The efficiency of the AEC electrolysis is currently similar to alkaline, i.e. in the range of 40-69% [43]. An advantage of the PEM electrolyser in comparison with alkaline is its ability to work more flexibly due to its shorter response time, which allows it to operate in a fluctuating energy system [12,43,44]. In contrast to alkaline and proton exchange membrane electrolysis, SOEC electrolysers operate at a higher temperature, between 600 and 1000°C, which allows high efficiencies, above 80%. This high efficiency is mainly due to the ability to supply energy with heat instead of electricity [43].

SOEC electrolysers can work in reversible operation mode, which means that they can function both as electrolysers and fuels cells. This is known as reversible solid oxide fuel cells (RSOFC) [12,43,45]. Additionally, SOEC electrolysers are also able to conduct co-electrolysis of H_2O and CO_2 producing syngas, which directly can be converted into various types of transport fuels [43]. A comparison of the three electrolysers SOEC, AEC and PEM is presented in Table 12-Table 14. The tables are based on a comprehensive literature study by [43] and data from [46] and [44]. The report by IRENA [44] does not contain specific data for the SOEC electrolyser due to its low maturity level, but predicts that it can be a game-changing technology [44].

Table 12. Comparison of AEC electrolysers, performance and costs. Source (The Danish Energy Agency, 2019), (Brynolf et al., 2018), (IRENA, 2018)

Technology	**AEC**								
Source	**[7]**				**[18]**			**[19]**	
	2015	**2020**	**2030**	**2050**	**2030**	**2018**	**2030**	**2017**	**2025**
Energy/technical data									
Typical plant size (MW)	10	10	10	10	0.5-50	1.1-5.3	4.9-8.6	-	-
- Input									
Electricity input (%)	100	100	100	100	-	-	-	-	-
Heat input (%)	0	0	0	0	-	-	-	-	-
- Output									
Hydrogen output (%)	61.2	63.6 (62-65)[a]	65.9	69.2 (66-70)[a]	~ 70	65 (43-69)[b]	69 (50-74)[b]	65	68
Heat output (%)	0	14	12	8	~ 5	-	-	-	-
Financial data									
Investment Cost (M€$_{2015}$ per MW)	1.07	0.60	0.55	0.50	0.7 (0.4-1)[b]	1.1 (0.6-2.6)[b]	0.7 (0.4-0.9)[b]	-	-
Fixed O&M (€ per MW/year)	53.5	30,000 (20,000-40,000)[a]	27.5	25,000 (17,500-35,000)[a]	-	-	-	-	-
O&M cost (% of investment cost)	-	-	-	-	2-3	2-5	2-5	-	-
Stack replacement cost (% of inv.)	-	-	-	-	Incl. O&M cost	50	-	-	-
Technology specific data									
Operation temperature (°C)	80	80	80	80		60-80	-		
Operation pressure (bar)	-	-	-	-	-	≥ 1	-	1	15
System life span (years)	25	25	25	25	10-20	25 (20-30)[b]	30	20	20
Stack lifetime (1000h)	-	-	-	-	-	< 90	75 (60-90)[b]	80	90
Regulation ability, ramp up (minutes)	8	8	0.5	0.5	-	-	-	0.2-20%/ second	-
Regulation ability, ramp down (minutes)	8	8	0.08	0.08	-	-	-	0.2-20%/ second	-
Start-up time (minutes)	-	-	-	-	-	Min. to hours	-	1-10	-
Transient operation (% of capacity)	-	-	-	-	-	20-150	-	15-100	-

a) Uncertainties associated with the specific projections.

b) Ranges across studies in the review by Brynolf et al. (2018)

Table 13. Comparison of PEM electrolysers, performance and costs

Technology	**PEM**							
Source	**[7]**				**[18]**		**[19]**	
	2015	**2020**	**2030**	**2050**	**2018**	**2030**	**2017**	**2025**
Energy/technical data								
Typical plant size (MW)	1	10	10	10	0.10-1.2	2.1-90	-	-
- Input								
Electricity input (%)	100	100	100	100	-	-	-	-
Heat input (%)	0	0	0	0	-	-	-	-
- Output								
Hydrogen output (%)	54	58 (55-60)a	62	67 (63-72)a	62 (40-69)b	69 (62-79)b	57	68
Heat output (%)	-	-	12	10	-	-	-	-
Financial data								
Investment Cost (M€2015 per MW)	1.9	1.1 (0.8-1.5)a	0.6	0.4 (0.2-0.8)a	2.4 (1.9-3.7)b	0.8 (0.3-1.3)b	-	-
Fixed O&M (€ per MW/year)	95	55,000 (40,000-75,000)a	30	20,000 (10,000-40,000)a	-	-	-	-
O&M cost (% of investment cost)	-	-	-	-	02-maj	02-maj	-	-
Stack replacement cost (% of inv.)	-	-	-	-	60	-	-	-
Technology specific data								
Operation temperature (°C)	67	80	85	90	50-80	-		
Operation pressure (bar)	-	-	-	-	> 100	-	30	60
System life span (years)	15	15	15	15	20 (10-30)b	30	20	20
Stack lifetime (1000h)	-	-	-	-	95 (90-100)b	62 (20-90)b	40	50
Regulation ability, ramp up (minutes)	1	0.03	0.01	0.01	-	-	100%/second	-
Regulation ability, ramp down (minutes)	0.02	0.02	0.02	0.02	-	-	100%/second	-
Start-up time (minutes)	5	0.5	0.15	0.15	Sec. to min.	-	0-5	-
Transient operation (% of capacity)	-	-	-	-	5-100	-	0-160	-

a) Uncertainties associated with the specific projections.
b) Ranges across studies in the review by Brynolf et al. (2018)

Table 14. Comparison of SOEC electrolysers, performance and costs

Technology	**SOEC**					
Source	**[7]**				**[18]**	
	2015	**2020**	**2030**	**2050**	**2018**	**2030**
Energy/technical data						
Typical plant size (MW)	0.25	1	15	50	-	0.5-50
- Input						
Electricity input (%)	85	85	85	85	-	-
Heat input (%)	15	15	15	15	-	-
- Output						
Hydrogen output (%)	68	76 (72-80)[a]	79	79 (75-83)[a]	-	~ 70
Heat output (%)	3	3	1.5	1.5	-	~ 5
Financial data						
Investment Cost (M€$_{2015}$ per MW)	-	2.20 (1.35-3.0)[a]	0.6	0.4 (0.25-1.5)[a]	-	0.7 (0.4-1)[b]
Fixed O&M (€ per MW/year)	-	66,000 (44,000-110,000)[a]	18	12,000 (8,000-20,000)[a]	-	-
O&M cost (% of investment cost)		-	-	-	-	2-3
Stack replacement cost (% of inv.)		-	-	-	-	Incl. O&M cost
Technology specific data						
Operation temperature (°C)	775	740	675	650	600-1000	
Operation pressure (bar)	-	-	-	-	-	-
System life span (years)	-	20	20	20	-	10-20
Stack lifetime (1000h)	-	~ 40	~ 60	~ 90	-	-
Regulation ability, ramp up (minutes)	1	1	1	1	-	-
Regulation ability, ramp down (minutes)	1	1	1	1	-	-
Start-up time (minutes)	60	60	-	-	-	-
Transient operation (% of capacity)	-	-	-	-	-	-

a) Uncertainties associated with the specific projections.

b) Ranges across studies in the review by Brynolf et al. (2018)

As shown in Table 12, the AEC has the lowest investment costs in comparison with SOEC and PEM, while its system lifetime is also longer. Due to the membranes and electrodes in the PEM electrolyser, which typically consist of noble metals such as iridium and platinum, this electrolyser has high capital costs [39,43]. In contrast, the SOEC electrolyser does not use any precious components and has a high potential for utilising the electrical energy, close to 100%, as it is able to reuse the waste heat from the electrolysis process. SOEC can therefore become a very cheap electrolysis technology [47]. The main advantages of the PEM electrolysis are its fast regulation ability and its responsiveness to load changes, which allow the PEM electrolyser to operate more flexibly [12]. The proton exchange membrane electrolysis is therefore favourable in an intermittent energy system [39]. The SOEC electrolyser is still in the development phase. However, a pilot project in Dresden by Sunfire has proven that SOEC can achieve an efficiency above 80% [48]. SOEC electrolysis offers the highest potential efficiency and can operate both in fuel cell and electrolysis mode (as described previously). This makes this technology very attractive in systems with fluctuating power production and improves the economic incentives due to its double function [49].

BIOGAS METHANATION

The methanation of biogas to electro-methane, which enables excess CO_2 to react with hydrogen from an electrolysis technology, can both take place in a biological process or catalytically through the conversion of syngas into methane and water [12,17,43]. In this process, the carbon oxides and dioxides are combined with hydrogen to create more molecules of CH_4. The methanation process is an exothermal process, and besides creating methane as an end-product, the process also releases heat. Depending on the type of methanation used, the heat is either low-grade or high-grade waste heat. The biological and catalytic methanation process will be explained below.

Catalytic methanation

Catalytic methanation has been known since the beginning of the last century, and several types of reactors exist: fixed-bed, fluidised-bed, three phase and structured, with the first two as the most established technologies. The reactors built around this concept operate at high temperatures, between 200 and 550°C and under pressures between 1 and 100 bar. The catalysts used in the methanation reaction may be Ni, Ru, Rh and Co, with Ni as the most optimal, even though it requires a high purity biogas/syngas. The trace components, such as sulphur in biogas/syngas, are poisonous for the Ni; thus, this type of reactor requires a more thorough cleaning process.

The methanation of biogas usually takes place over a nickel-based catalyst where only the CO_2 fraction of the biogas is methanised, while the existing fraction of CH_4 in the biogas remains unchanged [17]. The carbon dioxide content in the biogas reacts with hydrogen to form methane and water. The chemical reaction is shown in equation (1) below:

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O + \Delta H \quad (1)$$

The reaction is exothermic, meaning that heat is derived from the process, with ΔH = -164.9 kJ/mol. The methanation of biogas consists of two steps. A reverse water-gas shift reaction is first taking place, forming CO by reaction with CO_2 and hydrogen:

$$CO_2 + H_2 + \Delta H \rightarrow CO\ H_2O \quad (2)$$

This reaction requires a heat input and is endothermic with ΔH = +41.5 kJ/mol. The Sabatier reaction subsequently forms methane by reacting with CO and hydrogen:

$$CO + 3H_2 \rightarrow CH_4 + H_2O + \Delta H \quad (3)$$

This Sabatier reaction is carried out at temperatures between 250 and 400°C [45], and the reaction is exothermic with ΔH = -206.2 kJ/mol [17]. The main challenge with these types of reactors is temperature control, because of the highly exothermic reactions. The methanation reaction produces steam and this can be utilised with advantage in the district heating network, which typically has a temperature of 70-90°C, or as feedstock to a SOEC electrolysis to supply its requirement for water and heat inputs. The need for external energy input for the electrolyser can thereby be reduced. The heat can alternatively be used in the anaerobic digestion process, which is, however, less energy efficient. In order to maximize the overall energy efficiency of the methanation process, the utilisation of the excess heat is in any case important [17]. Another benefit of the high operating temperatures in the reactor is the faster reaction time enabled by a better hydrogen transfer compared to biological methanation, which only operates at temperatures of maximum 70°C [50].

Biological methanation

Biological methanation is a well-established process that has been known for more than 100 years. The process has, however, only recently reached the demonstration level [17]. In this type of reactor, the methanation process is caused by bio-catalysts through methanogenic microorganisms. The biological methanation can operate on pressure levels between 1 and 10 bars, but pressurized operation improves the performance.

For the biological methanogenesis of biogas a certain type of archaea microorganisms is used to produce CH_4 using CO_2 as a carbon source and hydrogen as a reducing agent. This process is to some extent similar to the catalytic biogas methanation process, but chemically these processes are different. The reaction is shown in the equation below:

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O + \Delta H \quad (4)$$

The excess heat from the reaction ΔH is -130.7 kJ/mol [37]. As the microbes only perform the conversion when CO_2 and hydrogen are available, biological methanation is a highly controllable process. The microbes are less sensitive to the inlet gases than the Sabatier process. However, if the level of oxygen is too high, it will harm the microbes. The temperature of the biological methanation is considerably lower than the Sabatier process, i.e. 40-70°C where most of the demonstration projects operate on temperature levels around 60°C. As the ratio between CO_2 and hydrogen is equal to the catalytical methanation process, additional hydrogen from an electrolyser is needed in order to obtain a full utilisation of carbon dioxide in the biogas [17].

The microbes for the production of methane can either be placed inside the biogas reactor (*in-situ*) or in a separate reactor (*ex-situ*) [17]. Although the in-situ technology has been known for more than 20 years, it is still being tested at pilot scale [38]. As an advantage of in-situ, the whole process of methanation takes place within one anaerobic digester, thus lowering the investment cost as there is no need for an additional reactor.

There are, however, practical challenges due to the limited solubility of hydrogen to water. By carefully injecting hydrogen into to the reactor and by extensive stirring, this problem can partly be overcome. However, this will have a negative impact on the efficiency as it will increase the electricity demand for the process. It is therefore difficult to achieve a high effectiveness with a methane output above 90% in an economic manner [17]. An alternative to in-situ and ex-situ is to combine these as a hybrid in the context of a full scale plant [51]. This alternative can provide a more favourable pathway compared to the individual system [52]. In Table 15, figures from four methanation plants are presented.

Table 15. Data sheet for different pilot biogas methanation plants in Denmark and Germany. Source [17]

	Unit	Foulum, DK-Catalytic methanation	Wertle, DE–Catalytic methanation	Allendorf, DE-Biological methanation	Avedøre, DK–Biological methanation
Operation start		2013	June, 2013	Spring, 2015	April, 2016
Electrolysis					
Electrolyser type		SOEC	AEC	PEM	AEC
Electrolyser input capacity	MW	0.05 (50kW)	6	0.30 (300 kW)	1.2
Hydrogen output	Nm^3/h	17.3	1310	60	200
Oxygen output	Nm^3/h	8.65	655	30	100
Operating temperature	°C	725	80	-	-
Operating pressure	bar	-	-	40	13
Regulation ability	% of max	-	30-100	-	4-100
Methanation					
Reactor type		Boiling water type	Fixed-bed	Separate reactor	Separate reactor
CO_2 source	-	Directly from biogas	Biogas CO_2 removal	From biogas plant	Wastewater treatment
Hydrogen input	Nm^3/h	17.3	1300	60	200
Biogas input	Nm^3/h	10	-	-	60
CO_2 input	Nm^3/h	4.3	325	15	25
Biomethane + e-methane output	Nm^3/h	10	325 (only e-methane)	15 (only e-methane)	60
Methane CH_4 content (measured)	%	97.7	>91	>95	>98
Operating pressure	bar	Medium pressure	8-10	5	9
Operating temperature	°C	280	250-550	40	63
Efficiency					
Electrolysis efficiency (LHV)	%	91	70 (HHV)	-	51
Methanation efficiency (LHV)	%	79.1	77	75-80	84
Useful heat output from methanation	%	14	10	-	15
Total efficiency (el. to methane + heat) LHV	%	90.3 (design value)	64	-	70
Costs					
Electrolyser investment	M€	n/a	n/a	n/a	2.1
Methanation investment	M€	n/a	n/a	n/a	1.75

**Figures from Wertle and Allendorf are design values.*

Haldor Topsøe coordinated a small-scale methanation plant in Foulum in collaboration with Aarhus University and other actors. The SOEC electrolyser has shown an efficiency of 91%, as the excess heat from the methanation is utilised as an input for the electrolyser. The design value of the efficiency was 96.5%, but it has not been reached as the full synergetic utilisation has not been implemented at the demonstration plant [17]. The methanation plant in Wertle is placed in connection to a large biogas plant that upgrades biogas to the gas grid. Thereby, the methanation plant does not receive biogas but only carbon dioxide from the upgrading process. The biological methanation plant in Allendorf is very flexible due to its proton electrolyser membrane and biological methanation and can be started and stopped in less than one minute. Also, Electrochaeas' biological methanation plant in Avedøre can be ramped up and down almost instantly by regulating the gas flows [17]. In Table 16, a comparison of costs and projections of methanation found in the literature is displayed. It is possible to compare the five sources on cost per produced mega-watt methane (M€). On this parameter, it is clear that the costs of MeGa-StoRE are significantly lower than the costs of the other sources and are based on the projection costs that take a point of departure in upscaling a small pilot-plant.

REGULATION ABILITIES

In the power-to-gas applications, flexibility is key. Thus, the dynamic operation of the methanation reactor can significantly reduce the costs for hydrogen storage, known as a one of the most expensive components of the electrofuel plant.

According to [50], all reactor designs presented above have the potential to be operated dynamically, but load changes are influenced by the design of the plant, the control process, and the peripheral equipment. At the current level of research, it is known that the flexible operation does not significantly influence the catalysts. In the case of biologic methanation, there is no minimum load for the biologic processes, and these respond well to fast ramping rates. Nevertheless, in regard to the overall operation of the electrofuel plant, the energy content of the produced CH_4 should not be lower than the energy consumed to keep the plant on stand-by. In the case of catalytic methanation, upward or downward regulation is possible, with a minimum load of 40% (or lower in some cases) for fixed-bed reactors and 10-20% for three-phase reactors.

The complete stop of both reactor concepts is also possible, and no negative effects have been observed in the case of biological methanation. For the catalytic reactor, the process is different, requiring the reactor to be flushed with hydrogen or other inert gases and kept at a temperature above 200°C (for fixed-bed reactors). For example, the three-phase reactor can keep the high temperature due to the liquid phase, requiring less energy [50].

Table 16. Comparison of different costs and projection of catalytic methanation.

a) Average of plant in Germany and Sweden

Source	[12]				[53]				[54]				[43]			[55]
	2015	2020	2030	2050	2015	2020	2035	2050	2015	2020	2030	2050	2020	2030	2050	2018
Energy/technical data																
Capacity (MW methane)	2.3	3.3	8.3	23.1	0.01	0.47	18.81	18.81	10	10	54	54	5	50	200	
Methane (Nm^3/hour)					1	43	1710	1710								
Methane (Nm^3/year)																
Methane (GJ/year)	70,000	100,000	250,000	700,000												
Methane (GWh/year)	252	360	900	2,520												
Heat generation (MW)					0.00	0.09	3.40	3.40								
Financial data																
Cost (€/Nm^3/h methane)					-	603	469	416								
Cost (€/GJ methane)	35.8	30	25.2	14.9	-	-	-	-	-	-	-	-	-	-	-	~ 29.3[a]
Total cost (M€/plant)	2.51	3.00	6.31	10.40		0.03	0.80	0.71	7	7	36	36	3.0	15	50	
Cost (M€/MW gas out)	1.09	0.91	0.76	0.45	-	0.064	0.043	0.038	0.70	0.70	0.67	0.67	0.6	0.3	0.2	1.5
Fixed O&M (€/GJ methane)	0.14	0.12	0.10	0.06												
Variable O&M (€/GJ SNG)	0.14	0.12	0.10	0.06												
O&M (% of investment)									2	2	2	2	4	4	4	
Technical specific data																
Lifetime (years)	25	25	25	25									25	25	25	19
Operating hours	8,000	8,000	8,000	8,000	-	-	-	-								8,760
Efficiency, methanation (%)													77	77	77	83

BIOGAS TO METHANOL, DME AND JET FUELS

Methanol, DME, diesel, petrol or jet fuels can be produced either from biogas/biomethane or the conversion of the separated CO_2 from biogas to these fuels (see Figure 11). There are a variety of different pathways to produce these fuels including different complexity levels. Methanol production from biogas can be performed by *direct conversion* via partial oxidation, photo-catalytic or biological conversion or by *indirect conversion* as biogas reforming to syngas and subsequent conversion to methanol [56]. The catalytic conversion of biomethane to syngas followed by either Fischer-Tropsch synthesis to desired alternative fuels (gasoline, diesel and jet fuel) or the fermentation of the generated syngas to different alcohols (methanol, ethanol) are the most dominant methods. The biological conversion of biogas to methanol seems to be a promising pathway due to its high conversion efficiency [56]. DME can be produced by tri-reforming of biogas [57] or by converting obtained methanol from biomethane. Boingartz *et al* [58] analysed the production of different transport fuels by converting green carbon dioxide with electrolytic hydrogen by CO_2 hydrogenation.

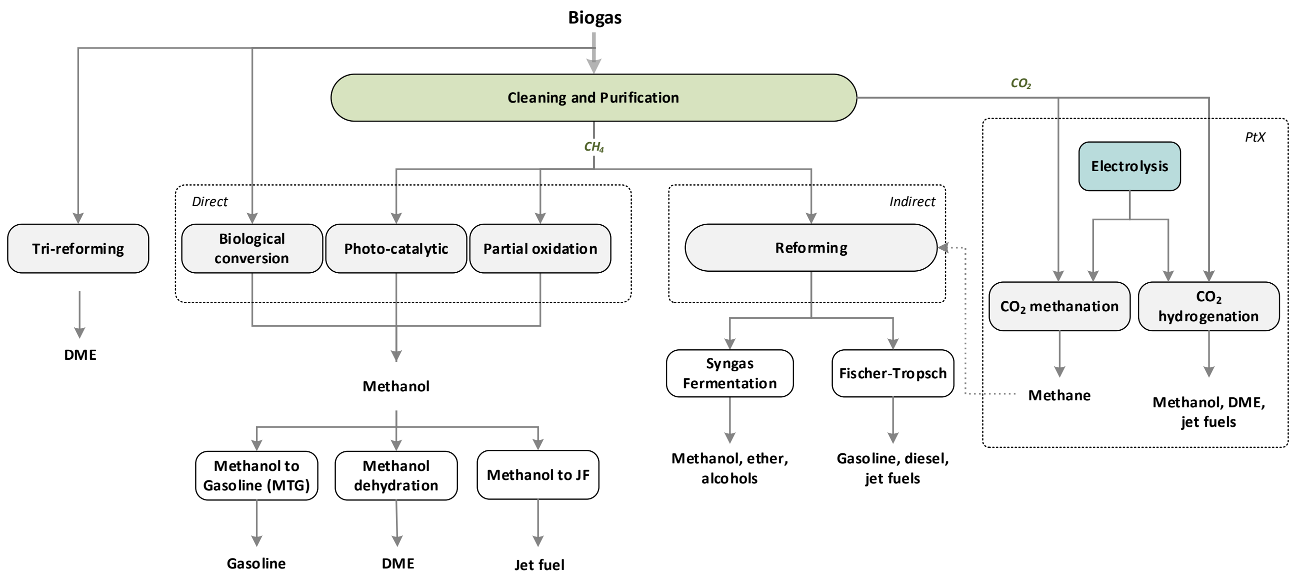

Figure 11. Different pathways of converting biogas to different end-fuels

The production of jet fuels can be obtained also with the addition of electrolytic hydrogen. In this case, the CO_2 yield of biogas is firstly methanated with the addition of hydrogen and, secondly, the pure methane (electro-methane) is further reformed to jet-fuels. The CO_2 hydrogenation process to methanol and its further conversion to jet-fuel is another possible way to produce aviation fuels [59]. These processes could also use CO_2 from biogas upgrading. Jet-fuel derived from methanol has good cold start properties and seem to be a promising alternative to the jet-fuel produced by the Fischer-Tropsch pathway [59].

Some of the conversion pathways are more mature; partial oxidation and the reforming of methane have been commercialized; biological conversion is being demonstrated, and photocatalytic methanation is currently only at the research level [56]. Syngas fermentation to ethanol has been commercialized, while the conversion to methanol and other alcohols is still at the research level. CO_2 hydrogenation to methanol over heterogeneous catalysts based on copper is the most mature technology [60,61]. A pilot plant for the conversion of biogas to jet-fuels with the addition of hydrogen is to be established in 2020 in Denmark as a part of the eFuel project [62].

INTEGRATION OF RENEWABLE ENERGY BY P2G VIA BIOGAS METHANATION

An energy system analysis of renewable energy integration via biogas methanation has been conducted by the use of the EnergyPLAN tool. The tool can simulate biogas methanation with the addition of electrolytic hydrogen and it shows the interaction of these technologies with the rest of the energy system. The EnergyPLAN tool operates on an hourly basis and can analyse the hourly fluctuations of renewable energy sources. For the analysis, it was chosen to look into a Danish reference scenario of 2020 [63] and the IDA Energy Vision scenarios for 2035 and 2050 [64]. All analyses were done by using the technical simulation in EnergyPLAN, which identifies the least fuel consuming system in each step. Table 17 summarizes key parameters used in the analysis.

Table 17. Key system parameters for the Danish energy systems for 2020, IDA 2035 and 2050

	Unit	Ref 2020	2035	2050
Demands				
Electricity	TWh/year	33.25	30.22	32.92
DH demand	TWh/year	29.92	30.51	28.19
Individual heating	TWh/year	20.46	15.72	14.51
Industry	TWh/year	25.73	20.39	8.32
Transport	TWh/year	60.05	43.15	32.85
Primary energy supply				
Wind (onshore & offshore)	TWh/year	19.05	39.7	70.85
Solar PV	TWh/year	1.01	4.26	6.35
River hydro	TWh/year	0.02	0	0
Wave	TWh/year	0	0.61	1.35
Coal	TWh/year	19.13	0.64	0
Oil	TWh/year	78.08	28.93	0
Natural Gas	TWh/year	25.15	18.39	0
Biomass	TWh/year	59.73	43.98	49.93
Excess electricity production	TWh/year	0.58	0.13	3.4
Conversion capacities				
Onshore wind	MW_e	4232	4000	5000
Offshore wind	MW_e	2051	6000	12000
PV	MW_e	952	3500	5000
River hydro	MW_e	3	0	0
Wave	MW_e	0	176	300
Large CHP	MW_e	1760	1926	3500
Small CHP	MW_e	876	1026	1500
Power plant	MW_e	1909	2574	3000
Large-scale heat pumps	MW_e	65	700	700
Electrolysis	MW_e	0	3547 (2948)*	6908 (5742)*

**In brackets electrolysis capacity without electrolysis for biogas methanation*

The scenario for 2050 was adjusted with a higher share of electrification in the transport sector, reducing the demand for liquid fuels by 12% to 24.29 TWh. The electrolysis tested in the reference 2020 scenario is the alkaline electrolyser as the only mature and commercially available technology, with an efficiency of 58.6% [12]. In the IDA 2035 and IDA 2050 scenarios, SOECs were tested with an efficiency of 74% [12]. Additional losses were added to the values from the DEA catalogue, including 5% losses due to the grid connection and 5% losses due to the hydrogen storage.

Maximal biogas potential assumed for the reference scenario in 2020 is 5.42 TWh, 7.15 TWh for 2035 scenario and 11.7 TWh for 2050 scenario. The levels of e-methane were varied from 0 to using maximal indicated biogas potential, by 7 steps, for all three scenarios as visualized in Figure 12. The efficiency of 82% for gas output per gas and hydrogen input was used to determine e-methane output, with share of hydrogen in the total gas input to the methanation unit of 37%.

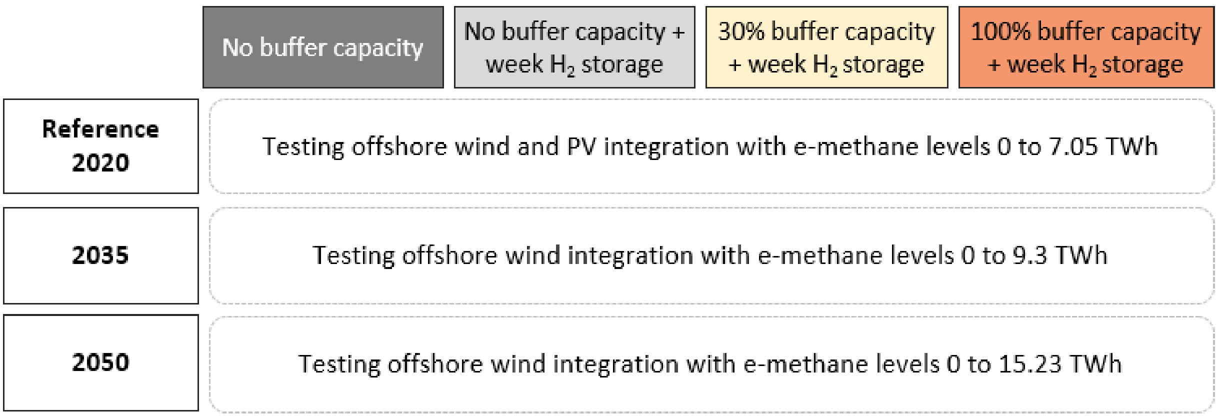

Figure 12. Overview of the simulated scenarios with electrolysis configuration, e-methane levels and renewable energy integrated

Different electrolysis set-ups were tested in order to identify the influence on the integration of renewable energy. Additional buffer capacities of 30% and 100%, respectively, were added to the electrolysis capacity to meet the hydrogen demand in a constant mode, and were supplied with one-week hydrogen storage in comparison with no storage at all. The electrolysis capacity and hydrogen storage were adjusted according to the hydrogen demand for biogas methanation.

In addition, electricity system price duration curve for high and low electricity prices were generated for 2020 and 2050 scenario with and without biogas methanation in the system to visualize the impact of implementation of e-methane on the electricity system price.

RENEWABLE ENERGY INTEGRATION IN THE 2020 REFERENCE SCENARIO

In the reference scenario for 2020, the potential for integration of intermittent electricity (different offshore wind and PV capacities) has been analysed. The test involved seven different levels of biogas methanation that can substitute the natural gas demand, from no biogas methanation to 7.05 TWh of produced methane, where all the available biogas used in the reference system was methanated. Figure 13 shows the electrolysis capacity with the different levels of e-methane in the system and for the different buffer added.

2020 Electrolysis capacity [MW]	E-methane in the system [TWh]						
	0	1.25	2.5	3.75	5	6.25	7.05
No buffer capacity	0	110	220	329	4382	548	618
30% buffer capacity	0	141	285	427	571	712	803
100% buffer capacity	0	218	439	657	878	1107	1236

Figure 13. Electrolysis capacity for different biogas methanation levels in 2020 scenario

Figure 14 illustrates the levels of integration of intermittent electricity in relation to different electrolysis configurations. The offshore wind capacity was changed in order to increase the share of renewable electricity in the system as more biogas methanation was included in the system. In the reference system, a natural gas demand of 25 TWh was therefore displaced by methanated biogas.

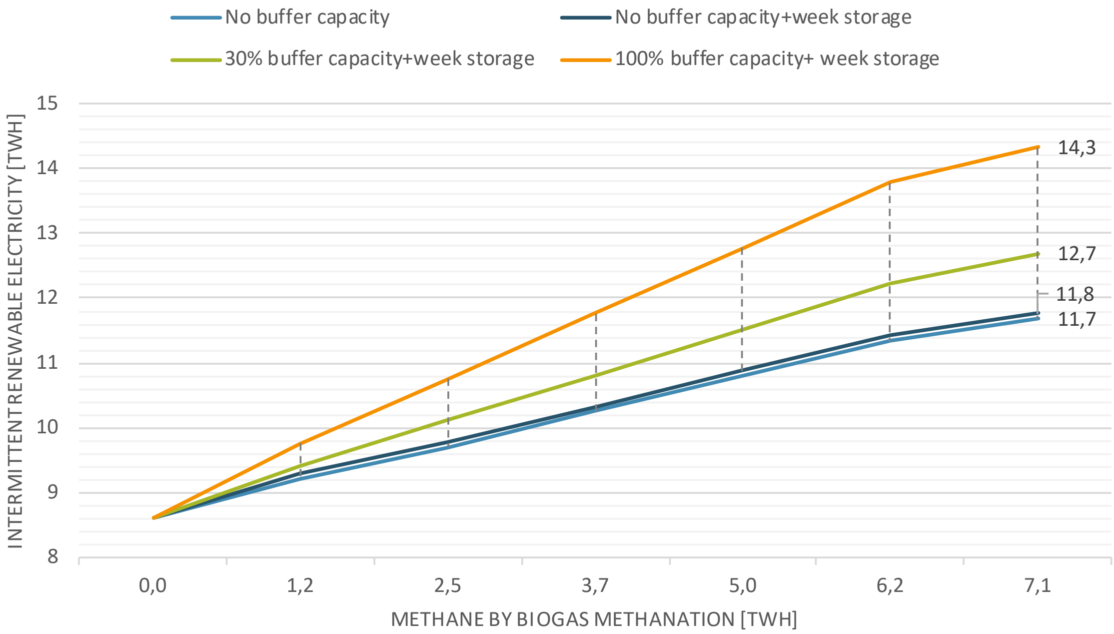

Figure 14. Integration of intermittent renewable electricity by offshore wind capacity changes via biogas methanation in the reference 2020 model

In the scenario in which a 100% buffer capacity and one week of hydrogen storage were added, the offshore wind capacity and the electricity produced increased by 22% in comparison with the constant operation of electrolysis for biogas methanation and no hydrogen storage. In this case, all biogas available in the system was methanated. Overall, installing biogas methanation with buffer capacity and storage resulted in an 11% increase in the total intermittent electricity share in the energy system. The penetration rates of intermittent electricity were higher when additional electrolysis capacity and storage were used.

On this basis, it can be concluded that the integration of a one-week hydrogen storage without buffer capacity allows a very small increase in the renewable electricity supply, while the increase of the buffer capacity to 100% including a one-week hydrogen storage has a bigger impact.

In Figure 15, the critical excess electricity production (CEEP[2]) for different electrolysis buffer, hydrogen storage and biogas methanation levels is investigated. Offshore wind capacities varied from 0 to 5000 MW, corresponding to 0-21.01 TWh. The figure illustrates how flexible these different scenarios are in terms of integrating intermittent electricity.

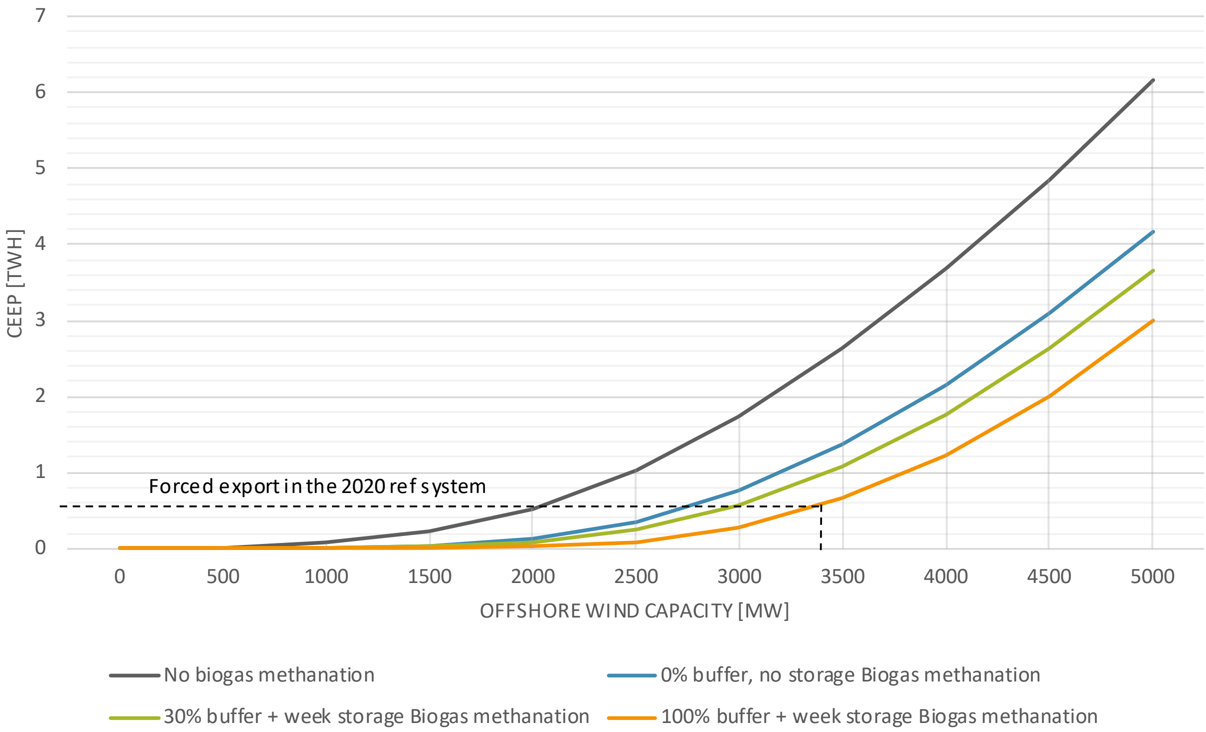

Figure 15. Critical excess electricity production in 2020 system for different levels of electrolysis capacity and hydrogen storage for biogas methanation and increasing offshore wind capacity

[2] Electricity that cannot be used in the system and needs to be exported.

It is visible from the figure that the system without biogas methanation can integrate up to 2051 MW of offshore wind, corresponding to the same forced export levels as the system with 100% electrolysis buffer and one-week hydrogen storage that can integrate 3410 MW. Therefore, 1359 MW more offshore wind can be integrated by installing electrolysis with buffer capacity and hydrogen, corresponding to an increase of 66% in comparison with the system without biogas methanation.

Increasing the electrolysis capacity helps the integration of intermittent electricity; however, it has a negative effect on the efficiency of the overall energy system. As we can see from Figure 16, increasing the electrolysis buffer and storage does reduce the primary energy supply in comparison with no additional capacity or storage. It is also visible that the lowest primary energy supply in the systems with biogas methanation can be identified at higher offshore wind capacities; however, the system without electrolysis shows an overall lower primary energy supply.

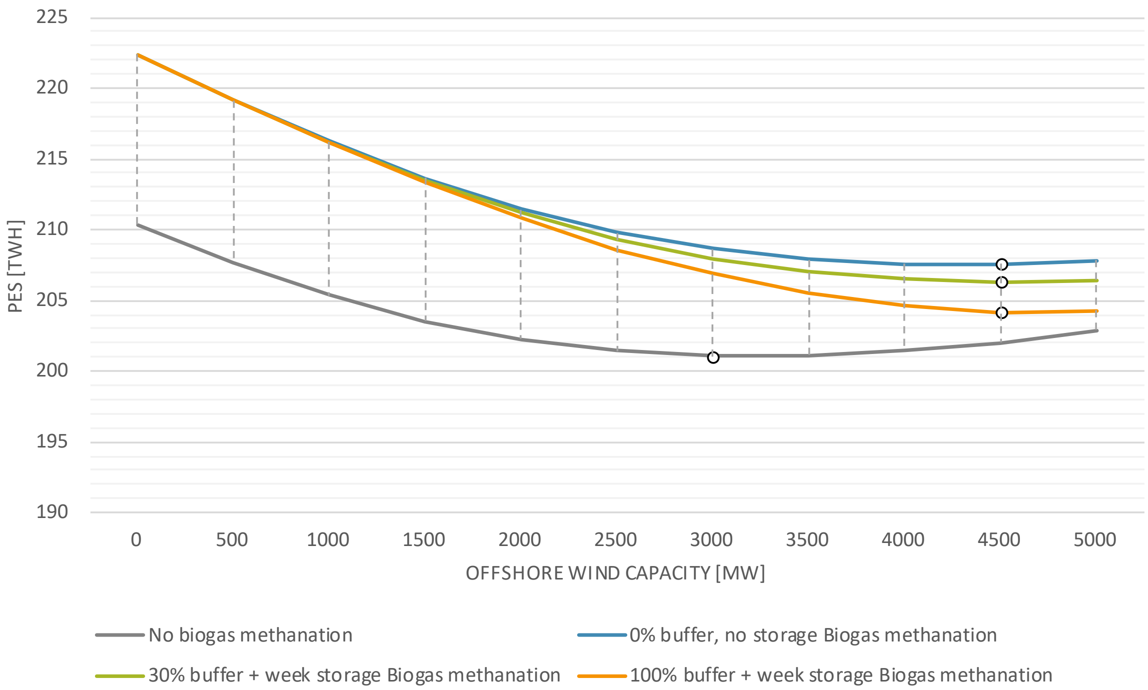

Figure 16. Primary energy supply in 2020 system for different levels of electrolysis capacity and hydrogen storage for biogas methanation and increasing offshore wind capacity

Figure 17 illustrates the levels of integration of intermittent electricity, where the PV capacity varied as more biogas methanation was included in the system. A similar trend can be seen as in the case of integrating offshore wind; however, the penetration rate is much higher in the case of integrating PV into the system. This can be attributed to the different characteristics of PV technology and its specific operation time in comparison to offshore wind. In the case of 100% buffer capacity and one week storage, the PV capacity has reached its maximum potential of 5000 MW in the last three steps and hereby offshore wind capacity was added to supplement the needed electricity.

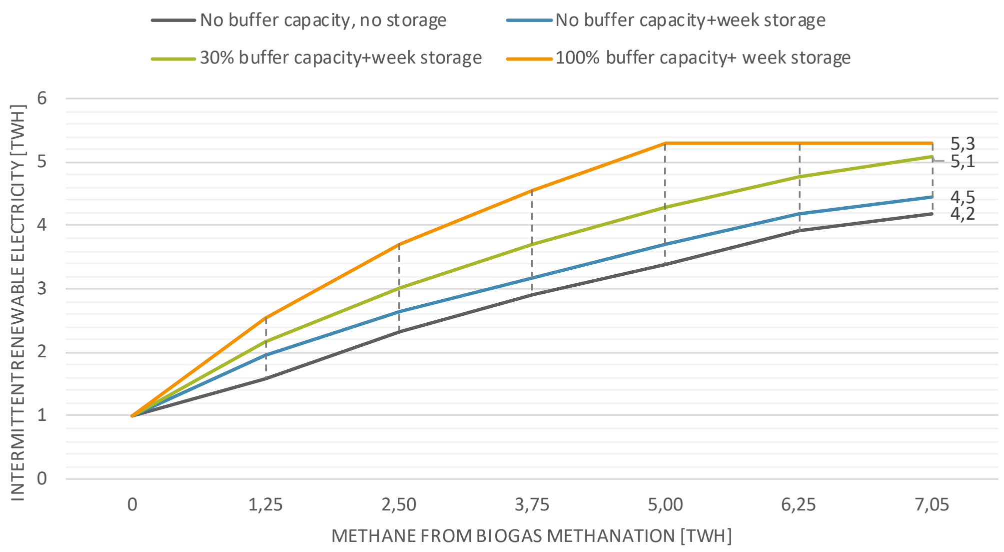

Figure 17. Integration of intermittent renewable electricity at PV capacity changes via biogas methanation in the reference 2020 model

PV capacities varied from 0 to 5000 MW, corresponding to 0 to 5.3 TWh. Figure 18 illustrates the flexibility of the different biogas methanation scenarios in terms of integrating intermittent electricity. It can be seen in the figure that the system without biogas methanation can integrate a maximum of 1000 MW of PV. This corresponds to the same forced export levels in comparison with the system with a 100% electrolysis buffer and a one-week hydrogen storage that could integrate 6000 MW of PV, which is higher than the PV potential in Denmark. Therefore, 5000 MW more PV can be integrated by installing electrolysis with a 100% buffer capacity and a one-week hydrogen storage.

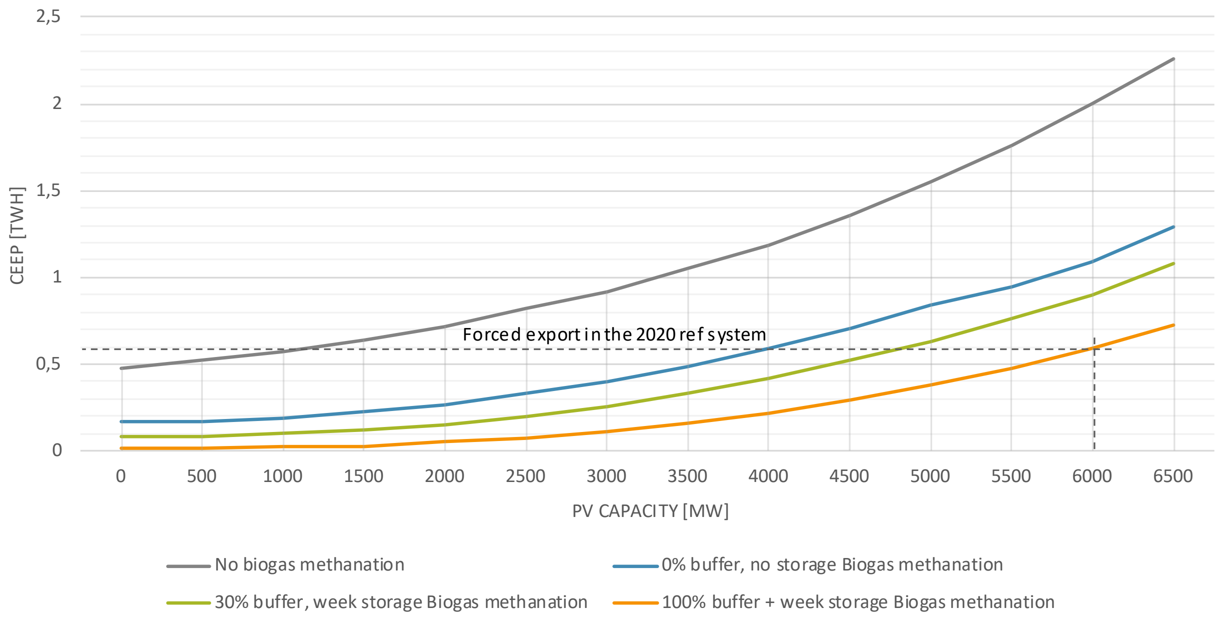

Figure 18. Critical excess electricity production in 2020 system for different levels of electrolysis capacity and hydrogen storage for biogas methanation and increasing PV capacity

Figure 19 illustrates the duration curves for the hourly electricity market price at the high electricity price level (77 €/MWh) and basic fuel price level (35 €/MWh), for both biogas purification and biogas methanation. The marked green area shows the effect of the biogas methanation on the electricity market price. We can see that biogas methanation increases the electricity system market price as it converts the electricity to hydrogen and thus uses more electricity than the system with biogas purification. The effect is in the range of 0-4 €/MWh.

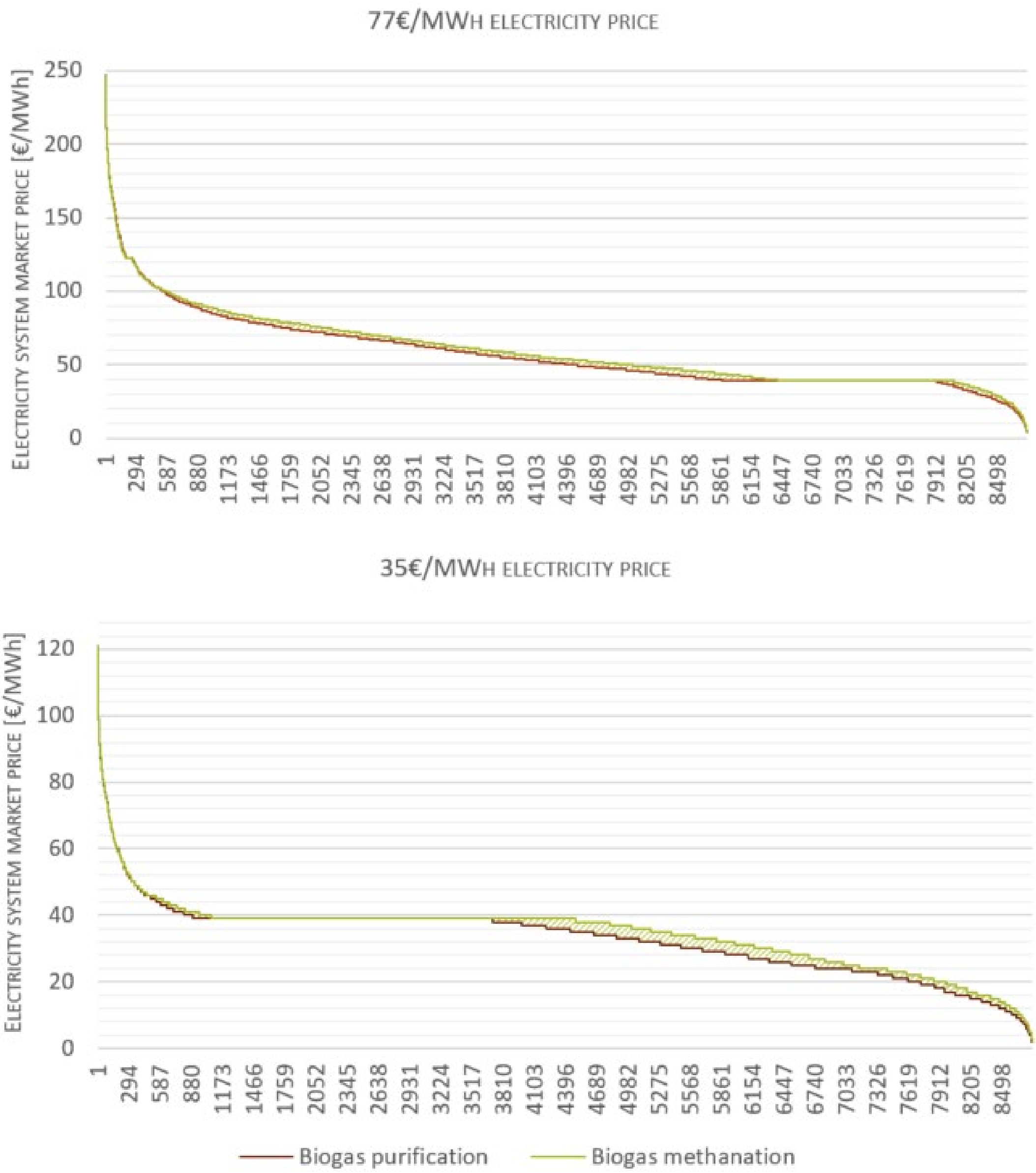

Figure 19. Electricity system price duration curve for high and low electricity prices for the reference 2020 model. The shaded areas represent the total effect of biogas methanation on the hourly electricity market price

RENEWABLE ENERGY INTEGRATION IN THE 2035 SCENARIO

In the IDA Energy Vision scenario for 2035, the demand for methanated biogas is 4.45 TWh. In the analysis, this value was changed from 0 to 9.3 TWh to see the ability of the system to integrate renewable sources by implementing P2G, where 9.3 TWh corresponds to methanating 7.15 TWh of biogas. The electrolysis capacity in this analysis varies from 2947 MW to maximum 4257 MW in the case of 100% buffer capacity and methanation of the full biogas potential (see Figure 20). Different levels of offshore wind capacity were used in order to increase the share of renewable electricity as more biogas methanation was included in the system.

2035 Electrolysis capacity [MW]	E-methane in the system [TWh]						
	0	1.43	2.86	4.45	5.75	7.15	9.3
No buffer capacity	2947	3048	3148	3260	3352	3451	3602
30% buffer capacity	2947	3079	3209	3354	3474	3602	3798
100% buffer capacity	2947	3150	3350	3574	3758	3955	4257

Figure 20. Electrolysis capacity for different biogas methanation levels in 2035 scenario

We can see a similar trend from Figure 21 as in the reference model, though the potential for integrating renewable energy is slightly smaller due to the already installed electrolysis capacity in the system (2947 MW) for liquid fuel production. The preinstalled electrolysis capacity has a 100% buffer included and has one-week storage (182 GWh). By adding extra capacity and one-week storage for biogas methanation, 11% more renewable electricity can be integrated than in the case of no additional capacity or storage. This was 22% more renewable electricity in the reference system. The conclusion is therefore the same; additional capacity and hydrogen storage improve the ability of the system to integrate more renewable electricity, but in the IDA 2035 scenario, this is limited due to the previously installed electrolysis capacity.

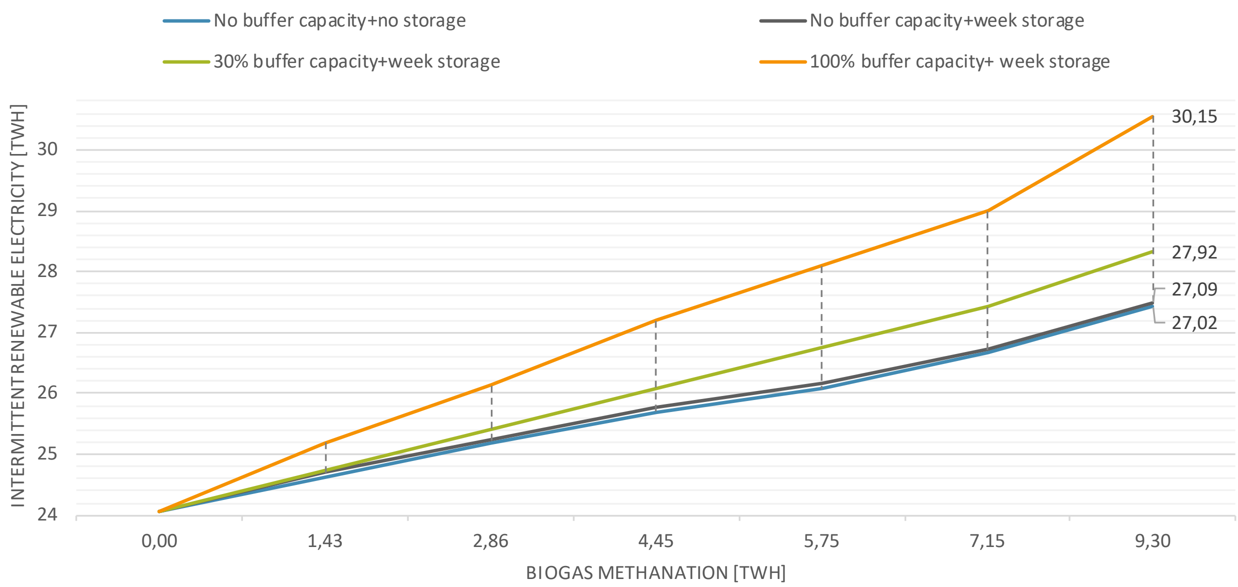

Figure 21. Integration of intermittent renewable electricity at offshore capacity changes via biogas methanation in IDA 2035

The offshore wind capacity varied from 0 to 16,000 MW, corresponding to 0 to 71.47 TWh. Figure 22 shows the critical excess electricity production, illustrating that in the case of additional electrolysis capacity and one week of hydrogen storage, the system can integrate 6750 MW of offshore wind in comparison with the system without biogas methanation, where it is possible to integrate 5295 MW to keep the same forced export of electricity.

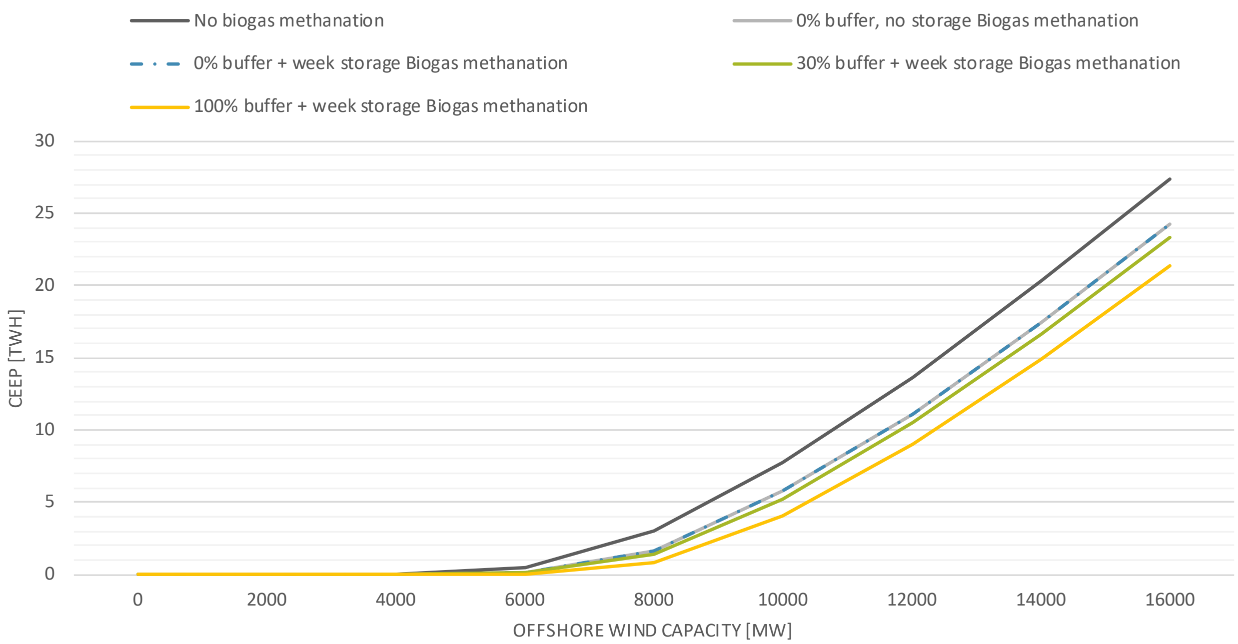

Figure 22. Critical excess electricity production in the 2035 system for different levels of electrolysis capacity and hydrogen storage for biogas methanation and increasing offshore wind capacity

RENEWABLE ENERGY INTEGRATION IN THE 2050 SCENARIO

The IDA Energy Vision scenario for 2050 includes an annual demand of 24.29 TWh for liquid electrofuels for heavy-duty transport and 8.41 TWh of methanated biogas. The 2050 system already includes 6908 MW of electrolysis and 432 GWh of hydrogen storage. In the analysis, this value was changed from 0 to 15.23 TWh to see the ability of the system to integrate renewable sources by implementing P2G, where 15.23 TWh corresponds to methanating 11.7 TWh of biogas. The electrolysis capacity varied from 5820 MW in the system without biogas methanation to a maximum of 7963 MW in the system with 100% buffer capacity (Figure 23).

2050 Electrolysis capacity [MW]	E-methane in the system [TWh]						
	0	2.34	4.68	7.02	9.36	11.7	15.23
No buffer capacity	5820	5985	6149	6314	6478	6643	6876
30% buffer capacity	5820	6035	6248	6463	6676	6890	7194
100% buffer capacity	5820	6151	6478	6801	7136	7467	7963

Figure 23. Electrolysis capacity for different biogas methanation levels in 2050 scenario

The analysis shows (Figure 24) that the potential for integrating renewable energy with biogas methanation in the system with an existing high electrolysis capacity is not as big as in the reference system. It is only possible to implement 9% more wind than in the case of no biogas methanation in comparison with the 22% in the 2020 reference system.

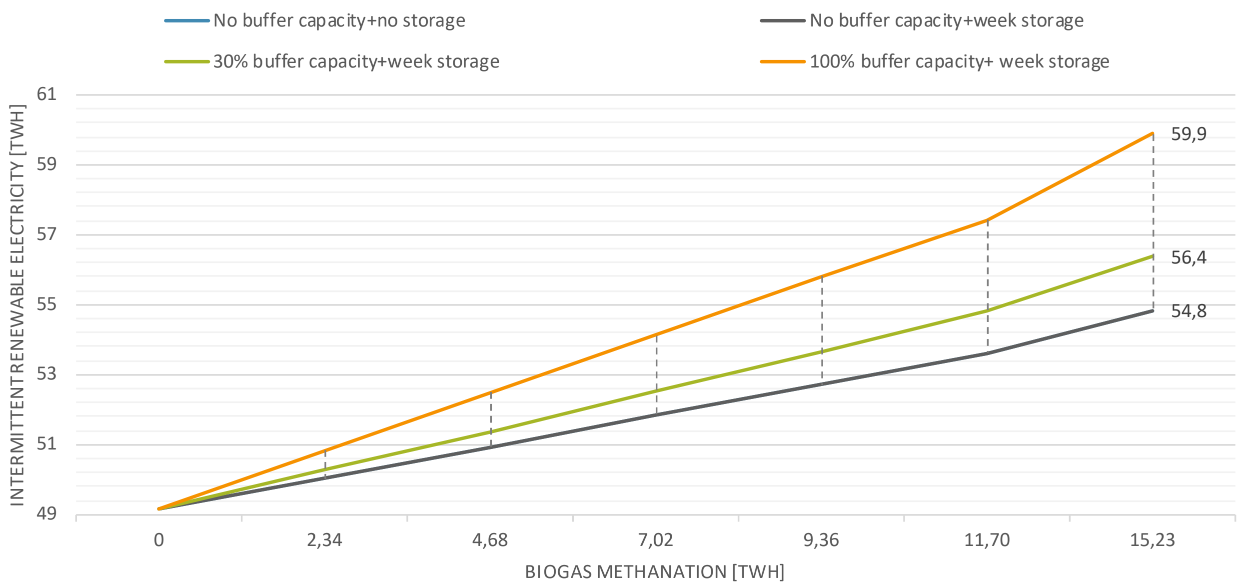

Figure 24. Integration of intermittent renewable electricity at offshore wind capacity changes via biogas methanation in 2050

By using a buffer capacity and a one-week storage, 22% renewable energy can be integrated, while in the case of no buffer with or without storage, it is possible to integrate 11.5% renewable energy. This is due to the existing overcapacity of electrolysis for the liquid electrofuel production; therefore the impact of the biogas methanation in this system is not that visible.

In order to test the flexibility of the system, the offshore wind capacity varied from 0 to 16,000 MW, corresponding to 0 to 71.47 TWh. By keeping the same forced export in the system with biogas methanation with buffer capacity and one-week storage, we can increase the installed offshore wind capacity from 10,790 MW to 13,150 MW in comparison with the system without biogas methanation (Figure 25).

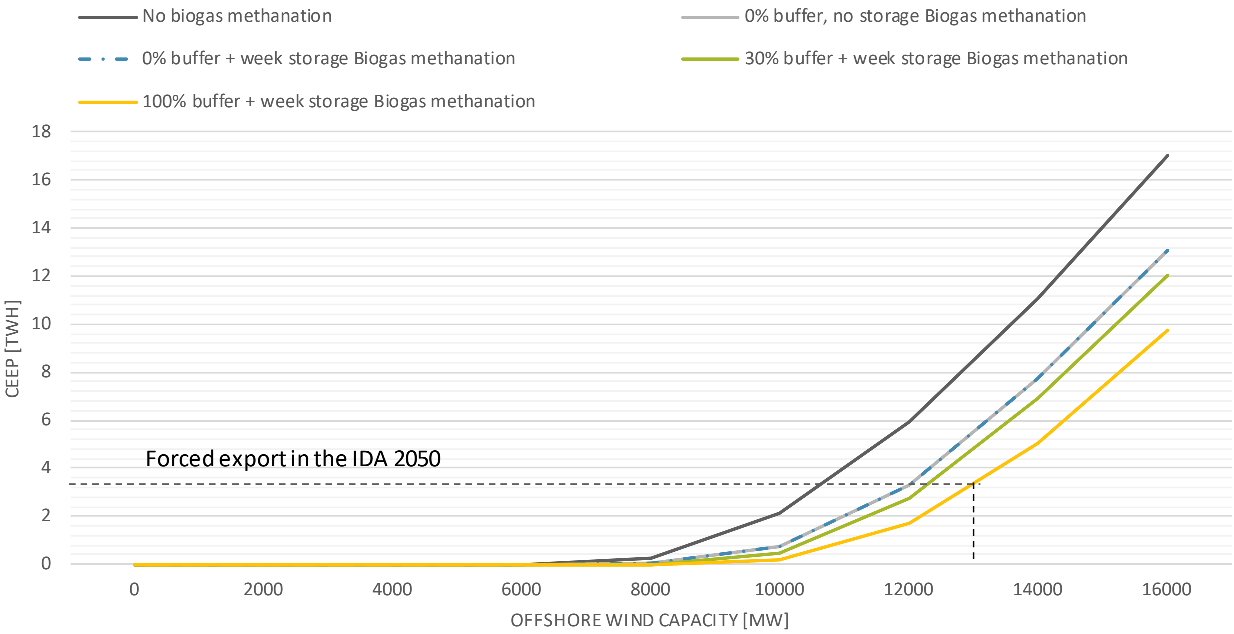

Figure 25. Critical excess electricity production in the 2050 system for different levels of electrolysis capacity and hydrogen storage for biogas methanation and increasing offshore wind capacity

Figure 26 illustrates the duration curves for the hourly electricity market price at the high electricity price level (77 €/MWh) and basic fuel price level (35 €/MWh), for both biogas purification and biogas methanation. The marked green area shows the effect of the biogas methanation on the electricity market price. We can see that biogas methanation increases the electricity system market price as it converts the electricity to hydrogen and thus uses more electricity than the system with biogas purification. The effect is in the range of 0-12 €/MWh in the case of the high electricity price and in the range of 0-6 €/MWh in the case of the low electricity price.

Figure 26. Electricity system price duration curve for high and low electricity prices for the IDA 2050 model. The shaded areas represent the total effect of biogas methanation on the hourly electricity market price

The analysis shows that if the biogas methanation is to play a role in the smart energy system, hydrogen storage and additional electrolysis capacity need to be installed. The integration of renewable energy is higher if the electrolysis is properly sized and if the storage is used; however, the results are more sensitive to the capacity of electrolysers rather than the installed storage. In addition, biogas methanation can provide heat for district heating and, depending on the implemented capacities in the future energy system, the plants should potentially be located close to the district heating grid.

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